THE PROCEEDINGS OF
THE PHYSICAL SOCIETY

Section A

Vol. 64, Part 7 1 July 1951 No. 379 A

Paramagnetic Resonance in Vanadous Ammonium Sulphate

By B. BLEANEY, D. J. E. INGRAM and H. E. D. SCOVIL
Clarendon Laboratory, Oxford

MS. received 11*th December* 1950

ABSTRACT. In the solid state the orbital momentum of the divalent vanadium ion $3d^3$ is quenched by the cubic crystalline field, leaving a spin quadruplet as the ground state. Paramagnetic resonance measurements show that in $V(NH_4)_2(SO_4)_2.6H_2O$ this is split into two doublets with a separation of 0·36 cm^{-1} by a component of the crystalline field with rhombic symmetry. An extensive and isotropic hyperfine structure is observed, from which the value of 7/2 for the nuclear spin of ^{51}V is confirmed. The contribution from the observed splittings to the tail of the magnetic specific heat curve is evaluated.

§ 1. INTRODUCTION

In previous work, the hyperfine structure of the paramagnetic resonance spectra of divalent ions of copper, cobalt and manganese has been studied. These ions are all members of the first (iron) transition group, and their stable isotopes are all odd, with strong nuclear magnetic moments. In this group there is one other such ion, divalent vanadium, of which the abundant isotope (99·8%) is of mass 51. The nuclear spin of this isotope has been determined from the optical hyperfine structure by Kopfermann and Rasmussen (1936) as 7/2, but this value is still given in some tables with a query. It seemed therefore of interest to examine the paramagnetic resonance spectrum to check this value, and to continue the survey of the iron group.

The results obtained in the work on copper, cobalt and manganese have been interpreted by Abragam and Pryce (1951). They have found that to explain the hyperfine structure it is necessary to refine the theory and to include the perturbing effect of excited levels which is not otherwise manifest in the behaviour of the electronic levels. The case most similar to vanadium is that of manganese, where the 3d shell is just half filled and the free ion is in a state $3s^2 3p^6 3d^5$, ^6S (neglecting inner closed shells). In this state the orbital momentum is zero, and the electron cloud distribution has spherical symmetry; it follows that the magnetic field at the nucleus has no contribution from either the orbit or the spin. (In this respect the effect of an S state formed from a half-filled shell of 3d-electrons is very different from that of an s-electron.) Thus there should be no observable hyperfine structure, but in fact a large hyperfine structure is found. This anomaly has been explained by Abragam (1950) as due to admixture by configurational interaction of an excited $3s\,3p^6 3d^5 4s$, ^6S state, where the presence of an unpaired s-electron produces a very wide hyperfine structure.

The free ion V^{++} is in a $^4F_{3/2}$ state, but the orbital momentum is strongly quenched by the crystalline electric field, as in the case of the isoelectronic ion, trivalent chromium. The orbital levels are split by a cubic field into two triplets and a singlet, the latter lying lowest, some 10,000 cm^{-1} below the nearest triplet. The wave function of the singlet orbital level is xyz, and it is fourfold degenerate in the spin. It may easily be verified that with this distribution the electronic spin moments produce no magnetic field at the nucleus. Thus the hyperfine structure would be zero, except for a small contribution from the orbital magnetic moment which is admixed by the spin–orbit coupling. It is to be expected, however, that the mechanism proposed by Abragam to account for the hyperfine structure in manganese, and also postulated in the theory of the cobalt and copper salts, should be operative in vanadium. Any hyperfine structure of appreciable magnitude must be assigned to this cause, which should give rise to an isotropic hyperfine structure.

If the symmetry of the crystalline electric field is lower than cubic, the degeneracy of the electron spin quadruplet is lifted. Neglecting nuclear effects, the quadruplet is split into two doublets. When a magnetic field is applied, the remaining twofold degeneracy is lifted, but even in strong fields the four levels are not equally separated (Figure 1). The spectrum consists, therefore, of three electronic transitions, of relative intensities 3 : 4 : 3. The central transition occurs at a field which is independent of the Stark splitting, and the others are symmetrically disposed about it, the separation being determined by the Stark splitting and the anisotropy of the crystalline field. Thus the latter, and the spectroscopic splitting factor g, can be found from the fine structure. This simple picture applies only to the case of very strong fields, but corrections can be applied for the deviations observed in finite fields.

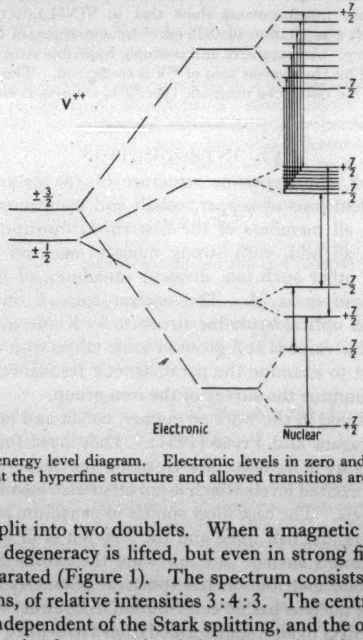


Figure 1. Schematic energy level diagram. Electronic levels in zero and strong magnetic field: on the right the hyperfine structure and allowed transitions are indicated.

The hyperfine structure results in a further splitting, and each electronic transition in strong fields is split into $(2I+1)$ equally separated components. The additional energy of each component due to the electronic–nuclear interaction is proportional to Mm, where M, m are respectively the electronic and nuclear magnetic quantum numbers. Thus the allowed transitions $M \rightarrow M \pm 1$, $\Delta m = 0$ result in a hyperfine structure of $(2I+1)$ components, with a constant spacing which is the same for all electronic transitions.

§ 2. THE EXPERIMENTS

The crystals used in these experiments were of vanadous ammonium sulphate, $V(NH_4)_2(SO_4)_2.6H_2O$, kindly prepared for us by Dr. P. F. D. Shaw. This salt crystallizes as a Tutton salt, and belongs to the series on which most of our previous measurements have been made. The V^{++} ion oxidizes rather easily, but is more stable in the form of a mixed crystal formed with the corresponding zinc salt ($V : Zn \simeq 1 : 1000$) as required to obtain narrow lines. The crystallization took place rather rapidly, so that the crystals were not as perfect as could be desired; however it was decided to proceed with these samples, rather than risk oxidation in attempting to recrystallize. The only feature of the spectrum which might be attributed to imperfect crystals is the anomalous line width of some transitions (see § 3.2). For details of the crystal structure of the Tutton salts the reader is referred to Bleaney, Penrose and Plumpton (1949).

The first experiments were performed at a wavelength of 3 cm. A large number of lines was obtained, some of them almost at zero field. The grouping indicated that there was an extensive hyperfine structure, together with a splitting of the electron spin quadruplet of the order of 0·3 cm^{-1}. To determine the nature of the crystalline field causing this splitting by examining the transitions in strong fields, it was necessary to use a shorter wavelength, and all the subsequent measurements were made at 1·2 cm. wavelength.

The experimental technique is exactly similar to that used in the case of manganese (Bleaney and Ingram 1951), the spectrum being finally obtained as a trace on an oscilloscope screen, from which it can be traced or photographed.

§ 3. RESULTS

It has been shown by Abragam and Pryce (1951) that the spectrum obtained in paramagnetic resonance may be interpreted in terms of a Hamiltonian

$$\mathscr{H} = g\beta \mathbf{H}.\mathbf{S} + D\{S_z^2 - \tfrac{1}{3}S(S+1)\} + E\{S_x^2 - S_y^2\} + AS_zI_z + B(S_xI_x + S_yI_y). \qquad \ldots\ldots(1)$$

Here g is assumed isotropic, which is found to be the case. The term in D represents the fine structure due to field of axial symmetry, while departures from this due to a rhombic component appear as the term in E. In manganese the latter is small compared with D, and this was assumed to be likely in vanadium also. The separation between successive fine structure lines would then vary as $(3\cos^2\theta - 1)$ when a strong field is applied at an angle θ with the z axis. It turned out that the value of E for vanadium was large, so that this simple variation with angle did not hold.

The hyperfine structure due to magnetic interaction with the nucleus is represented by terms in A and B. Owing to the closely cubic symmetry of the electron cloud, the gradient of the electric field at the nucleus should be very small, and the electric quadrupole interaction negligible. The direct effect of the external magnetic field on the nucleus does not affect the spectrum when only transitions involving no change in the nuclear orientation are observed. The terms which would represent these effects have therefore been omitted from the Hamiltonian.

3.1. *The Fine Structure*

The investigation of the fine structure was based on the same principles as in the case of manganese. The Tutton salts have two ions in unit cell, the one being derived from the other by a reflection in the crystallographic *ac* plane. Two of the principal magnetic axes of the crystal (K_1, K_2) lie in this plane, the third (K_3) being perpendicular to it, along the *b* axis. The positions of K_1 and K_2 are determined by plotting the spectrum as the external magnetic field is applied at various angles in the *ac* plane, where the spectra of the two ions are identical. In Figure 2 the fields at which the three electronic transitions occur corresponding to $\Delta M = \pm 1$ (M being the electronic magnetic quantum number in strong fields) are shown as a function of ψ, the angle between the field and the *c* axis. The separation between the transitions rises to a maximum in two mutually perpendicular directions, which are therefore K_1 and K_2. The greater maximum lies at $\psi = +2°$, which is taken as K_1.

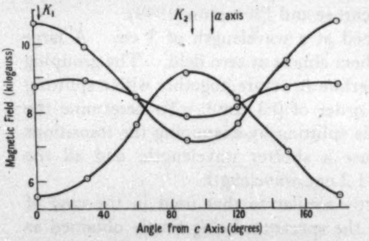


Figure 2. Magnetic fields at which strong absorption lines occur in the *ac* plane at $\bar{\nu}=0{\cdot}805$ cm^{-1}.

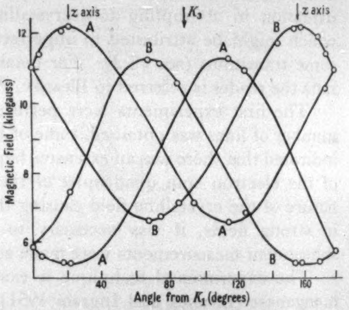


Figure 3. Magnetic fields at which the transitions $M=+\frac{3}{2}\rightarrow+\frac{1}{2}$ and $M=-\frac{1}{2}\rightarrow-\frac{3}{2}$ occur in the K_1K_3 plane at $\bar{\nu}=0{\cdot}815$ cm^{-1}. A, B refer to the two ions in unit cell.

To carry the analysis further, it is necessary to make some assumption concerning the behaviour of the two individual ions. Normally each is found to have nearly tetragonal symmetry in the Tutton salts; the tetragonal axes must then lie in either the K_1K_3 or the K_2K_3 plane. The separation of the fine structure lines should increase to a maximum in one of these planes, the direction of this maximum being the tetragonal axis of one ion; no such maximum should be observed in the other plane.

In Figure 3 are shown the positions of the two extreme outside transitions for each ion in the K_1K_3 plane as a function of the angle from K_1. The maximum splitting occurs at 22° from K_1, which then defines a tetragonal axis on the assumptions made hitherto. This splitting should decrease to zero and rise to a subsidiary maximum at right angles to the tetragonal axis. This maximum should be just half as great as that at 22°, and should be the same as that along K_2. This is quite obviously not the case, showing that there is a considerable rhombic asymmetry. This should cause the splitting along K_2 to fall below the half value as much as that the subsidiary maximum in the K_1K_3 rises above it, since these two directions are mutually perpendicular. This is found to be the case.

If the direction of maximum splitting in the K_1K_3 plane is taken as the z axis of the Hamiltonian (1), the values of D and E may be evaluated from the spectrum observed in this direction. The three electronic transitions (including nuclear effects) should lie at

$$h\nu = G + 2D - \frac{3E^2D}{(h\nu)^2} + Am + \frac{B^2}{2h\nu}\{I(I+1) - m^2 + 2m\},$$

$$h\nu = G \qquad + \frac{3E^2}{h\nu} \quad + Am + \frac{B^2}{2h\nu}\{I(I+1) - m^2\},$$

$$h\nu = G - 2D - \frac{3E^2D}{(h\nu)^2} + Am + \frac{B^2}{2h\nu}\{I(I+1) - m^2 - 2m\},$$

where $G = g\beta H$. At 1·27 cm. wavelength, and 20° K., they occur at fields of 5,345, 8,630 and 12,110 gauss respectively. Hence one finds $D = 1{,}730$ gauss, $E = 530$ gauss. At 90° K. the values are found to be $D = 1{,}720$ gauss, $E = 540$ gauss.

3.2. *Hyperfine Structure*

At 1·2 cm. wavelength the various electronic transitions are well separated when the magnetic field is oriented in a suitable direction, and the hyperfine structure of each is completely resolved. In Figure 4 photographs of the oscilloscope trace for two electronic transitions are shown. In each case eight lines are clearly seen, confirming the value 7/2 of the nuclear spin. The line width is different in the two cases, being narrower for the transition $M = \frac{1}{2}$ to $-\frac{1}{2}$, whose position is approximately independent of the Stark splitting. Since the width due to the random magnetic field of other ions should be the same for all lines it seems natural to attribute the discrepancy to a variation in the Stark splitting in different parts of the crystal. Any extra width due to this should disappear in directions where the fine structure collapses, but this appeared not to be the case. Thus there seems no obvious explanation of this line width difference, which is similar to, though more pronounced than, that observed in manganese (Bleaney and Ingram 1951).

The width of the hyperfine structure is found to be 670 gauss, and completely isotropic. The values of A, B are therefore 96 gauss. The second order effects in the hyperfine structure which were observed and analysed in the manganese spectra at 3 cm. wavelengths, are of smaller value here since they vary inversely as the field at which the lines occur, and the analysis of the vanadous spectrum was undertaken from the 1·2 cm. wavelength measurements. It was difficult to measure the individual splittings in the hyperfine structure to an accuracy of more than 3 gauss, since in most cases they occurred in high fields where the homogeneity and stability of the magnetic field are not too good. The calculated second order change in the separation of successive hyperfine structure lines is only 1·1 gauss.

3.3. *The Spectroscopic Splitting Factor g*

The g value was determined by using an organic free-radical marker, as described in the paper on manganese. This essentially consists of a measurement of the field separation between the 'g marker' and the centre of the electronic transition $M = +\frac{1}{2} \rightarrow -\frac{1}{2}$. This is done directly on the oscilloscope screen, and can be obtained with an accuracy of within ±2 gauss. This field position must

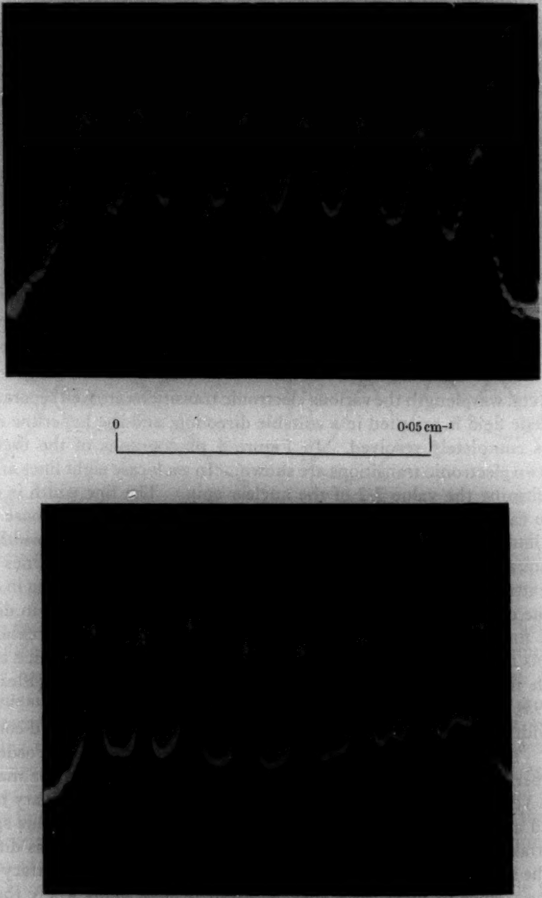


Figure 4. Photographs of hyperfine structure in electronic transitions $M=\frac{3}{2} \rightarrow \frac{1}{2}$ (above) and $M=\frac{1}{2} \rightarrow -\frac{1}{2}$ (below).

be corrected for the effect of second order terms in the splitting; for example, along the z axis of an ion the central transition * has an energy of

$$G+3E^2/G+31B^2/4G$$

instead of just G. Since the experimental error in E is relatively large, and E itself is considerable, this limits the accuracy of the g value obtained, and our measurements give a value of $1{\cdot}951 \pm 0{\cdot}002$ along the z axis.

So far as it was possible to ascertain, the g value remains isotropic, though at directions off the axis the second order corrections are larger, and the pattern more complicated to follow.

§ 4. DISCUSSION

Using the value of g just obtained, the values of the splitting parameters may be given in cm^{-1}. They are $A=B=0{\cdot}0088 \pm 0{\cdot}0002\ \text{cm}^{-1}$, $D=0{\cdot}158 \pm 0{\cdot}01\ \text{cm}^{-1}$, $E=0{\cdot}049 \pm 0{\cdot}005\ \text{cm}^{-1}$.

Here a rather large error has been given to D and E for the following reason. When the rhombic asymmetry is rather great, the K_1 axis is not necessarily the projection of the true ionic z axis on the *ac* plane, as is the case with tetragonal symmetry. There may thus be some error in the position derived for the z axis in our measurements, though as it lies at rather a small angle from the *ac* plane it should not be far out. The effect of such an error would be to diminish the apparent value of D and increase that of E.

It may be remarked that the ratio E/D is so large that there is little point in singling out the z axis in writing the Hamiltonian, as is done in (1) for near tetragonal symmetry. If $D=3E$, which is not far from our case, the Hamiltonian could be written $E'(S_z^2-S_y^2)$, with $E'=2D/3$. One would speak then only of a rhombic component, with no tetragonal component.

The contribution of these splittings to the specific heat can be evaluated from the expression (Bleaney 1950)

$$\frac{CT^2}{R}=\left(\frac{hc}{k}\right)^2\left\{\frac{1}{45}(D^2+3E^2)S(S+1)(2S-1)(2S+3)\right.$$
$$\left.+\frac{1}{9}(A^2+2B^2)S(S+1)I(I+1)\right\}.$$

The terms in D and E give a contribution of 0·067 and those in A and B of 0·0032, a total of 0·070. This value may be compared with the measurements of Eisses, Groendijk and Gorter (1940) from which a figure of 0·103 can be evaluated. The difference between these two results suggests that there may be a considerable effect due to exchange interaction in the concentrated salt, since the specific heat due to dipole–dipole interaction would amount only to $CT^2/R=0{\cdot}003$, leaving a discrepancy of 0·03.

From the data given above, a number of points of interest may be made from a comparison with the theory of Schlapp and Penney (1932). In a cubic crystalline field the orbital levels lie at 0 (triplet), $-8\,\overline{Dq}$ (triplet) and $-18\,\overline{Dq}$ (singlet). The value of $\overline{Dq}$ may be deduced from the g value, for $g=2(1-2\lambda/5\overline{Dq})$. For V^{++} the spin–orbit coupling constant $\lambda=55\ \text{cm}^{-1}$ (Laporte 1928); hence $\overline{Dq}=850\ \text{cm}^{-1}$. The singlet lies therefore at $-15{,}000\ \text{cm}^{-1}$ and transitions between it and the upper triplet will give rise to absorption extending into the visible region (the salt has a purple colour).

---

* The value given corresponds to the point half-way between the two central hyperfine lines.

To attempt to evaluate the splittings of the orbital triplets from the values of D, E would be very laborious, but a comparison may be made with the isoelectronic ion trivalent chromium. In the chrome alums the Stark splitting amounts to $0{\cdot}1-0{\cdot}2\,\mathrm{cm}^{-1}$, with trigonal symmetry. Van Vleck (1939) has shown that this is due to the splitting of the lower triplet, which is connected with the orbital singlet by spin–orbit coupling. The Stark splitting is of the order $\lambda^2 T/(\overline{Dq})^2$, if the trigonal field T is small compared with the cubic field. A small tetragonal or rhombic field (T, R) produces no first order splitting of the triplet, however, and the Stark splitting of the spin levels would be of the order $\lambda^2(T, R)^2/(\overline{Dq})^3$, which is an order of magnitude smaller than for the trigonal field. Thus the presence of a splitting in the vanadium Tutton salt which is rather larger than in the chrome alums shows that the rhombic field splitting of the orbital levels must be large, and probably comparable with the cubic splitting.

Pryce (1950) has calculated the magnitude of the electron spin–spin interaction which gives a small contribution to the electronic splitting in addition to the mechanism considered above. He finds that the magnitude of ρ in the Hamiltonian

$$\mathscr{H} = -\rho\{(\mathbf{L}\,.\,\mathbf{S})^2 + \tfrac{1}{2}(\mathbf{L}\,.\,\mathbf{S}) - \tfrac{1}{3}LS(L+1)(S+1)\}$$

is about $0{\cdot}4\,\mathrm{cm}^{-1}$, using spectroscopic data. This Hamiltonian has no diagonal elements within the ground state except through admixtures from higher states through spin–orbit coupling. The corresponding splitting of the spin quadruplet would therefore be of the order $\rho(\lambda/10\overline{Dq})$ or less than $0{\cdot}01\,\mathrm{cm}^{-1}$.

ACKNOWLEDGMENTS

We wish to acknowledge maintenance grants from the Department of Scientific and Industrial Research (D.J.E.I.) and the National Research Council of Canada (H.E.D.S.).

REFERENCES

ABRAGAM, A., 1950, *Phys. Rev.* **79,** 534.

ABRAGAM, A., and PRYCE, M. H. L., 1951, *Proc. Roy. Soc.* A, **205,** 135, **206,** 164, 173.

BLEANEY, B., 1950, *Phys. Rev.*, **78,** 214.

BLEANEY, B., and INGRAM, D. J. E., 1951, *Proc. Roy. Soc.* A, **205,** 336.

BLEANEY, B., PENROSE, R. P., and PLUMPTON, B. I. P., 1949, *Proc. Roy. Soc.* A, **198,** 406.

EISSES, J., GROENDIJK, H., and GORTER, C. J., 1940, *Physica,* **7,** 865.

KOPFERMANN, H., and RASMUSSEN, E., 1936, *Z. Phys.*, **98,** 624.

LAPORTE, O., 1928, *Z. Phys.*, **47,** 761.

PRYCE, M. H. L., 1950, *Phys. Rev.*, **80,** 1106.

SCHLAPP, R., and PENNEY, W. G., 1932, *Phys. Rev.*, **42,** 666.

VAN VLECK, J. H., 1939, *J. Chem. Phys.*, **7,** 61.

Secondary Elastic Scattering in Electron Diffraction

By J. M. COWLEY, A. L. G. REES and J. A. SPINK*
Division of Industrial Chemistry, Commonwealth Scientific and Industrial Research Organization, Melbourne, Australia

MS. received 18*th July* 1950, *and in amended form* 17*th January* 1951

ABSTRACT. Electron diffraction patterns from films of long-chain paraffins show the effects of secondary elastic scattering of electrons in the form of extra spots, groups of spots, arcs of circles, diffuse bands with sharp edges, and forbidden reflections in single-crystal patterns. Examples of such effects are given and analysed to demonstrate their origin in the successive diffraction of electrons from two different single-crystal regions. It is pointed out that many effects recorded in the literature on electron diffraction may be explained in the same way. The modification of the intensities in single-crystal patterns and the appearance of forbidden reflections are treated quantitatively on the basis of secondary diffraction by parallel but non-coherent regions of crystal. Expressions are derived whereby observed intensities may be corrected for secondary scattering when once the scattering efficiency of the crystal has been deduced from the observed intensities of forbidden reflections. The importance of such corrections for purposes of structure analysis is demonstrated in the comparison of two Fourier projections made from a cross-grating pattern of dicetyl ($C_{32}H_{66}$), one using observed intensities and the other using corrected intensities. Both show clearly the general arrangement of the molecules, but that based on corrected intensities contains much less spurious detail than the other, and gives greater accuracy and a much clearer indication of the positions of the hydrogen atoms.

---

§ 1. INTRODUCTION

ELASTIC scattering of electrons by crystals of suitable dimensions leads to the appearance of large proportions of the incident energy in the diffraction pattern. Consequently one might expect secondary elastic scattering of diffracted beams by underlying crystallites or mosaic elements of the same crystallite to contribute appreciably to the recorded electron diffraction pattern. In the literature on electron diffraction this process has been recognized as the origin of a number of the many spurious effects observed in electron diffraction patterns. 'Forbidden' reflections in single-crystal patterns have been explained in this way by Raether (1932), Taylor-Jones (1934) and other authors (Finch and Wilman 1937). Extra spots, given by successive reflections in different crystallites, have been reported by Darbyshire and Cooper (1935) for mica. In composite films extra spots are given by successive reflections in two or more layers of different structure, as observed by Finch and Sun (1936) for Cu-on-Pt and Ni-on-Pt films, and by Wilman (1940) for silver plus silver halide films. When the electron beam passes first through a single crystal and then a polycrystalline aggregate, arcs or rings occur about strong single-crystal reflections. This has been reported by Kirchner (1932) for CdI_2 on mica, and by Finch and Fordham (1937) for normal long-chain hydrocarbons.

Many other effects, such as extra spots and rings, groups of spots, diffuse bands with sharp edges and intensity anomalies have been reported, for example by Finch and Wilman (1936, 1937). It is the purpose of this paper to illustrate the occurrence of such effects in paraffin patterns and to show how secondary scattering may produce them. The way in which the relative intensities of reflections are modified by secondary scattering, and the method for correcting observed intensities to provide intensity data suitable for structure analysis by Fourier methods are outlined.

---

* Division of Tribophysics, C.S.I.R.O.

§ 2. EXPERIMENTAL METHODS

The investigations reported here were confined mainly to long-chain paraffin hydrocarbons and their mixtures, which crystallize in the orthorhombic system with the [001] axis perpendicular to the supporting film. By exploration of many specimens, prepared by evaporation of benzene solutions of these compounds, it was possible to find patterns illustrating the whole range of secondary scattering phenomena.

The possibility of observing much of the detail of these patterns rests in high resolution in the electron diffraction camera. Throughout these studies, the high-resolution camera designed by Cowley and Rees was used. High stability of the electrical supplies for the high tension and focusing coils, and the use of two $20\,\mu$ limiting apertures in the electron-optical system are the factors responsible for this high resolution. The accelerating voltage was 42 kv., and for most patterns the camera length was 54 cm.

For pure cross-grating patterns the intensity measurements required for comparison with theory were made with a Leeds and Northrop microphotometer, employing a square aperture of side 0·05 cm. The widths of reflections measured at a constant density from the microphotometer records were used to interpolate the intensities on a calibration curve of width against intensity, obtained from a series of exposures of the direct beam.

§ 3. TYPES OF PATTERN ATTRIBUTABLE TO SECONDARY DIFFRACTION

The main features of the various patterns obtained are illustrated in Figures 1–6 (Plates I–IV)*, which will now be described and interpreted.

(i) *Extra Spots*

In many transmission patterns from paraffin hydrocarbons there occur extra reflections lying between legitimate rings, particularly when only a limited number of random crystallites are contributing to the pattern (see Figure 1).

These extra spots can be completely interpreted on the basis of secondary elastic scattering. In Figure 1 the pattern has been completely analysed by identifying the five intense primary cross-grating patterns, and indexing all non-legitimate spots produced by secondary diffraction about the intense reflections of the primary patterns. The secondary cross-gratings, centred on the reflections of one primary cross-grating, lie in all cases in orientations parallel to one of the other primary cross-gratings, indicating that the crystallites are arranged in a stack with (001) planes parallel. It is possible in principle to identify the order in which the crystallites are stacked by consideration of the intensities of the secondary reflections, since equation (2) (see § 4), describing these intensities, is not symmetrical with respect to the subscripts 1 and 2. This arises from the curvature of the Ewald sphere. Identification on this basis depends on very accurate intensity measurement and does not seem to be practicable at present. One would expect to find only the strongest reflections in the secondary gratings, i.e. the set 110, $\bar{1}10$, $1\bar{1}0$, $\bar{1}\bar{1}0$, 200, $\bar{2}00$, 020 and $0\bar{2}0$.

---

* For Plates see end of issue.

The angles between the primary cross-gratings, and the assignment of the secondary spots are given in Table 1.

The primary cross-gratings are labelled A, B, C, D, E in order of diminishing intensity. The assignment of the origins of the secondary gratings has been made for one possible stacking order, namely, A, C, B, E, D.

The overlying transparency to Figure 1 shows the origin of all the extra spots in one quadrant of the pattern. An extra spot arising from secondary diffraction by grating B around an intense spot of the primary grating C is denoted C_B. In the transparency an example of an individual secondary cross-grating is distinguished. This is made up of A_B spots produced when the 110 spot of the A grating acts as origin of a secondary B grating. The relation between the spots of this secondary cross-grating is further emphasized by the close similarity of the fine structure, due to the mosaic nature of the crystals concerned in each. Since the spots of cross-grating B are less extended that those of A, most of the extension of the secondary spot groups is derived from that of the A spot acting as origin, and is parallel to this extension. Similarly the fine structure of other secondary spots may be explained in detail by consideration of the form of the primary spots.

Table 1

Angles between primary cross-gratings (°)		Origin of secondary grating	Angle measured (°)
A and B	53	A → B	53
C, C′	65, 67·5	A → C, C′	65·5, 68
D	32	A → D	32·5
E	86·5		
B and C, C′	59·5, 62	C, C′ → B	59·5, 62
D	84·5	B → D	84·5
E	32·5	B → E	33
C, C′ and D	34, 36	C, C′ → D	34, 36
E	26, 28	C, C′ → E	26, 28
D and E	62	E → D	62

Thus the detail of Figure 1 may be completely and unambiguously explained as the result of secondary scattering in a series of crystals. Similar processes may give rise to patterns containing just a few isolated non-legitimate spots or an almost continuous background of faint spots, depending on the structure of the specimen.

(ii) *Groups of Spots*

In patterns from mosaic crystals in which the mosaic blocks differ in orientation by small amounts, the extra spots due to secondary diffraction frequently form symmetrical groups about the primary cross-grating spots. Thus in Figure 1 the strongest cross-grating A is given by a group of crystals of slightly differing orientations. Figure 2 (*a*) is an enlargement of part of this pattern, showing the fine structure of the 020 reflection. Figure 2 (*b*) shows how this fine structure is produced by secondary scattering. The beam first passes through a group of small, slightly disorientated crystals, giving rise to the primary diffraction spot groups shown at the 020, 110, $\bar{1}10$, $\bar{2}00$ and 200 reflection positions.

These primary reflections are then diffracted by a large underlying single crystal, which gives a primary pattern at the points indicated by the heavier spots and the secondary spots about the 020 position as observed.

Very regular groups of spots are shown in Figure 3, an enlargement of part of a pattern obtained from polycrystalline dicetyl ($C_{32}H_{66}$). These groups reproduce in miniature a single crystal cross-grating, and may be attributed to secondary diffraction in crystals varying in orientation by intervals of about three degrees. A similar pattern was observed by Finch and Wilman (1937) from n-hexadecyl alcohol, but the explanation given by von Laue for this pattern was in terms of a superlattice of very large unit-cell dimensions (Finch and Wilman 1937). The occurrence of layers of crystal varying by a few degrees in orientation appears to be an example of the 'rotational slip' recently described by Wilman (1950).

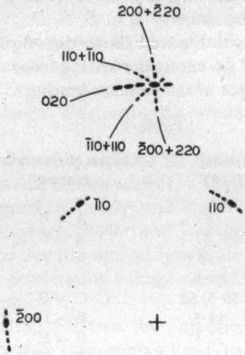


Figure 2 (*b*). Schematic diagram showing origin of spot group in Figure 2 (*a*).

(iii) *Forbidden Reflections in Single-Crystal Patterns*

Crystals of paraffins and other layer-lattice materials may give single-crystal patterns and yet contain discontinuities, particularly in the direction of the c-spacing, which would serve to render the scattering from various regions of the crystal incoherent. Hence a diffracted beam from one region could again be diffracted by a second region. In this way the intensities of single-crystal patterns may be considerably modified, and, in particular, reflections forbidden by the space-group symmetry of the crystal may be produced by the combination of two or more permitted reflections, independent of any dynamic interaction which may occur in perfect crystal regions.

For paraffin crystals the $h00$ and $0k0$ reflections with h or k odd are forbidden by the crystal symmetry, but are clearly seen in cross-grating patterns such as that reproduced in Figure 4. If a crystal giving such a cross-grating pattern is tilted about the [100] or [010] direction, the permitted reflections which could add up to give one of these forbidden reflections are not present, and it is observed that the forbidden reflections do not occur. This observation precludes the possibility that the forbidden reflections are produced by a modification of the structure.

In a few cases cross-grating patterns have been obtained similar in appearance to that of Figure 4, but with the forbidden reflections missing. Such patterns may be given by crystals in which there are relatively few discontinuities, so that little secondary diffraction takes place. If dynamic interaction in perfect-crystal regions were important for paraffins, it would have its maximum effect in such near-perfect crystals.

It is, as yet, difficult to assess the relative importance of secondary scattering and dynamic interaction in the production of the many forbidden spots reported in the literature. However, it seems probable that, at least for such poorly crystallized layer-type structures as the paraffins, secondary scattering must be the predominating effect.

(iv) *Secondary Segments and Complete Circles*

From specimens more polycrystalline than those considered above, the patterns contain very large numbers of secondary spots which form almost continuous segments of circles or complete circles about strong primary diffraction spots. Thus the pattern of Figure 5 was obtained from a large single crystal followed by some very fine crystalline material. Each strong primary spot is seen to be the centre of an almost complete 110 secondary ring, with some traces of a weaker 200 ring also present. The pattern reproduced in Figure 6 was given by many medium-sized crystals, and it is seen that the background is thickly covered with spotty rings, mostly centred at strong spots on the intense 110 and 200 rings.

(v) *Extra Rings and Bands*

The secondary diffraction spots lying on rings of radius r_2 about points on a primary diffraction ring of radius r_1 will form a family with envelopes, consisting of circles about the origin of radius (r_1+r_2) and (r_1-r_2). Considering only the strong 110 and 200 reflections of a paraffin pattern, the secondary circles will have three inner and three outer envelopes. The three inner envelopes will not usually be detected, being lost in the high-intensity region of the central spot. Of the three outer envelopes, two will coincide with the primary 220 and 400 rings. The third, however, with radius $(r_{110}+r_{200})$, will coincide with no primary ring, and so will give the effect of an extra ring. The formation of this extra ring as the envelope of the family of secondary circles is shown clearly in Figure 6. Faint extra rings have also been detected inside the 110 ring at radii equal to $(r_{020}-r_{110})$ and $(r_{020}-r_{200})$.

From specimens of still smaller crystals, giving continuous primary rings, this envelope will appear as a continuous extra ring. The remainder of the secondary rings will merge into a continuous background with intensity increasing from the primary rings out to the envelopes. Thus the effect of secondary scattering on continuous ring patterns will be to add a series of diffuse bands with intensity increasing to sharp maxima at extra rings or second-order rings. The microphotometer trace, Figure 7, shows the extra ring between the strong 220 and 400 rings of a paraffin pattern. The slow rise of the 'band' on the low-angle side and sharp fall-off on the high-angle side are evident. Such bands, corresponding to the outer envelopes, are readily distinguishable from those given by random two-dimensional lattices, which rise sharply and then fall off slowly as the radius increases. In combination, the two types of band could give some interesting effects. Thus, if the head of a two-dimensional band fell just outside the head of a secondary diffraction band, the intervening space would appear as a 'dark' ring or band. Finch and Wilman (1937) and others have observed a number of light and dark bands in patterns from metal films, mica, etc. Secondary diffraction effects are capable of giving such bands, and it is possible that some may originate in this way.

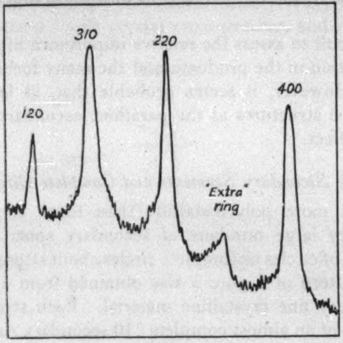


Figure 7. Microphotometer trace of paraffin wax ring pattern illustrating the occurrence and form of an 'extra' ring.

§ 4. THEORY OF SECONDARY SCATTERING IN SINGLE CRYSTALS

It has been demonstrated that secondary scattering may have marked effects on the intensity distribution and may also give rise to forbidden spots in single-crystal cross-grating patterns, such as those given by thin films of paraffins. Since for structure analysis it is desirable to have quantitative estimates of intensities of diffraction spots, it is necessary to treat secondary scattering quantitatively. The general treatment of this problem is somewhat cumbersome. Here we shall consider only the special case in which we are particularly interested, that of a cross-grating pattern given by transmission through a very thin crystal. For this case we make the approximation that the Ewald sphere has infinite radius, i.e. that all primary diffracted rays are parallel to the primary beam, and hence all secondary cross-gratings have the same intensity distribution as the primary cross-grating. This approximation is good when the crystal dimension is small in the direction of the beam, so that the reciprocal-lattice points are considerably elongated in this direction.

We consider the crystal to be made up of n equal layers, in each of which an electron beam is reduced in intensity by a fraction f by elastic scattering and a fraction g by inelastic scattering. After the beam has passed through r layers, the intensity of the h_1k_10 reflection of the $hk0$ cross-grating is $I^r_{h_1k_1}$. Of this, ${}^0I^r_{h_1k_1}$ is the intensity scattered out of the primary beam in the rth layer. For the first layer, ${}^0I^1_{h_1k_1} = I^1_{h_1k_1}$. The fraction of the intensity falling on any layer which goes into the $hk0$ reflection is given by $fI^1_{h_1k_1}/\sum_h\sum_k I^1_{hk}$. After passing through the n layers, $I^n_{h_1k_1}$ is made up of beams scattered once, twice, three times, etc. Those scattered only once give a contribution

$$\sum_{r=1}^{n} {}^0I^r_{h_1k_1}(1-f-g)^{n-r} = \sum_{r=1}^{n} I^1_{h_1k_1}(1-f-g)^{n-1} = n(1-f-g)^{n-1}I^1_{h_1k_1},$$

since the intensity of the primary beam entering the rth layer is

$$ {}^{0}I^{r-1}{}_{0} = I^{1}{}_{0}(1-f-g)^{r-1}. $$

A beam scattered twice is first scattered to h_2k_20 in the rth layer and then scattered to h_3k_30 in any of the $(n-r)$ succeeding layers, and the indices of the two reflections are related by $h_2+h_3=h_1$ and $k_2+k_3=k_1$. Hence the total contribution of beams scattered twice is

$$ \sum_{r=1}^{n-1}\left[\sum_{h_2}\sum_{k_2} {}^{0}I^{r}{}_{h_2k_2}\,.\,\frac{fI^{1}{}_{(h_1+\bar{h}_2)(k_1+\bar{k}_2)}}{\sum\limits_{h}\sum\limits_{k} I^{1}{}_{hk}}(1-f-g)^{n-r-1}(n-r)\right] $$

$$ = \sum_{r=1}^{n-1}\left[(n-r)f(1-f-g)^{n-2}\sum_{h_2}\sum_{k_2}\frac{I^{1}{}_{h_2k_2}\,I^{1}{}_{(h_1+\bar{h}_2)(k_1+\bar{k}_2)}}{\sum\limits_{h}\sum\limits_{k} I^{1}{}_{hk}}\right] $$

$$ = f(1-f-g)^{n-2}\,\frac{n(n-1)}{2}\,\frac{S^{1}{}_{1}}{\Sigma^{1}}. $$

From beams scattered three times the contribution is

$$ f^{2}(1-f-g)^{n-3}\,\frac{n(n-1)(n-2)}{3}\,\frac{S^{1}{}_{1,2}}{(\Sigma^{1})^{2}}, $$

where
$$ S^{1}{}_{1,2} = \sum_{h_2}\sum_{k_2}\sum_{h_3}\sum_{k_3} I^{1}{}_{h_2k_2}I^{1}{}_{h_3k_3}I^{1}{}_{(h_1+\bar{h}_2+\bar{h}_3)(k_1+\bar{k}_2+\bar{k}_3)}. $$

Then, for large n,

$$ I^{n}{}_{h_1k_1} = nI^{1}{}_{h_1k_1}(1-f-g)^{n-1} + \frac{n^{2}}{2}\frac{S^{1}{}_{1}}{\Sigma^{1}}f(1-f-g)^{n-2} + \frac{n^{3}}{3}\frac{S^{1}{}_{1,2}}{(\Sigma^{1})^{2}}f^{2}(1-f-g)^{n-3} + \dots . $$

The intensity which would be observed in the absence of multiple scattering is written $I^{*}{}_{h_1k_1} = nI^{1}{}_{h_1k_1}$. Analogously we write $S^{*}{}_{1} = nS^{1}{}_{1}$, $S^{*}{}_{1,2} = n^{2}S^{1}{}_{1,2}$ and $\Sigma^{*} = n\Sigma^{1}$. If f_0 and g_0 are the fractions of the intensity scattered per unit length and l is the length of the crystal, $f = f_0l/n$ and $g = g_0l/n$. We can write, for the observed intensity,

$$ I^{n}{}_{hk} = e^{-l(f_0+g_0)}\left[I^{*}{}_{hk} + \frac{f_0l}{2}\frac{S^{*}{}_{1}}{\Sigma^{*}} + \frac{(f_0l)^{2}}{3}\frac{S^{*}{}_{1,2}}{(\Sigma^{*})^{2}} + \dots\right]. $$

Considering only secondary scatterings,

$$ K.I^{n}{}_{hk} = I_{hk}{}^{*} + \tfrac{1}{2}f_0lS^{*}{}_{1}/\Sigma^{*}, \qquad \dots\dots(1) $$

where K is a constant. For forbidden reflections the theoretical intensity $I^{*}{}_{hk}=0$, so that $KI^{n}{}_{hk} = \tfrac{1}{2}f_0lS^{*}{}_{1}/\Sigma^{*}$.

These relations were tested for a dicetyl cross-grating pattern. Theoretical intensities were calculated from the structure found by Bunn (1939) for long-chain paraffins. These calculated intensities were then modified assuming various values for f_0l in equation (1). Figure 8 shows the calculated intensities, uncorrected and corrected for secondary scattering for the case $\tfrac{1}{2}f_0l = 0{\cdot}40$, plotted against observed intensities. In each case the 110 intensities are taken as equal. It is evident that the correction for secondary scattering gives a marked improvement in the agreement of observed and calculated values. There is seen to be a slight over-correction for the weaker reflections, corresponding to spots distant from the origin. This is the nature of the error to be expected from the assumption of infinite radius for the Ewald sphere. It can be shown that, if the curvature of the sphere is taken into account, the products of intensities in the summation S_1 should be written

$$I_1 I_2 f\left(\frac{\lambda}{2d_1^2}\right) f\left(\frac{\lambda}{2d_2^2} - \frac{\lambda}{d_1 d_2}\cos\phi_{12}\right), \qquad \ldots\ldots(2)$$

where d_1 and d_2 are the interplanar spacings of the sets of planes concerned, ϕ_{12} is the angle between then, and $f(z)$ is the function representing the variation of the scattering power around a reciprocal-lattice point in the direction of the beam.

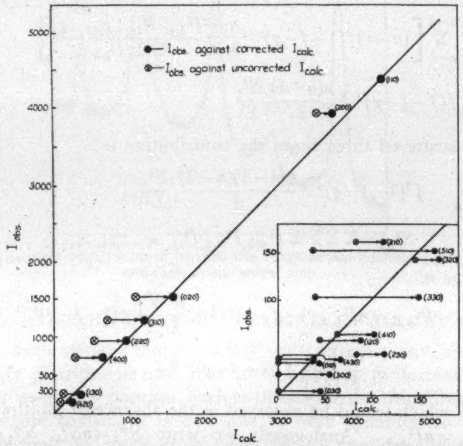


Figure 8. Comparison of observed intensities of single-crystal reflections from dicetyl with calculated intensities, uncorrected and corrected for secondary scattering.

For purposes of structure analysis it is necessary to reverse the above process and find the theoretical intensities I^*_{hk} from the observed intensities I^n_{hk}. Considering only secondary scattering, and neglecting terms higher than the second order, the inverse relation may be written

$$I^*_{hk} = K[I^n_{hk} - \tfrac{1}{2} f_0 l (S^n_1/\Sigma^n)], \qquad \ldots\ldots(3)$$

where the summations refer to observed intensities.

§ 5. APPLICATION TO STRUCTURE ANALYSIS

In order to assess the importance of the correction for secondary scattering in structure analysis, Fourier projections were made from the *hk*0 cross-grating of a dicetyl ($C_{32}H_{66}$) single crystal using first the observed intensities and then those corrected by the use of the formula (3).

Fourier projections obtained from electron diffraction data are projections of the potential distribution in the lattice, since the potential is given by

$$\Phi(xyz) = \sum_h \sum_k \sum_l E_{hkl} \exp\{-2\pi i(hx+ky+lz)\}$$

where E_{hkl} is the structure factor for electron scattering. However, because of the difficulties of obtaining significant electron diffraction intensity measurements, the Fourier method has been used only in isolated cases (e.g. Vanshstein and Pinsker 1949, 1950).

The intensities of the dicetyl pattern were deduced from spot half-widths, and no great accuracy can be claimed for them. The structure factors were taken as the square roots of the intensities. The measurements were limited to $hk0$ reflections with h from 0 to 7 and k from 0 to 6. The phases of the reflections were calculated by assuming the carbon atom positions found by x-ray diffraction. The forbidden reflections gave the value $\frac{1}{2}f_0l=0{\cdot}49$ in equation (3), and this value was used to correct the remainder. The values of the structure factors observed and corrected are listed in Table 2, together with the calculated signs.

Table 2

hk	Phase $hk0$	Phase $hk0$	F_{obs}	F_{corr}
10			10	0
20	+	+	79	73
30			10	0
40	+	+	34	20
50			8	0
60	−	−	9	6
01			10	0
11	+	+	100	100
21	−	+	16	15
31	+	+	45	31
41	−	+	12	5
51	+	+	16	0
61	−	+	8	4
02	+	+	80	76
12	−	+	18	16
22	+	+	44	46
32	−	+	18	16
42	+	+	18	0
52	−	+	11	9
62	−	−	8	4
72	−	+	5	4
03			10	0
13	+	+	34	28
23	−	+	17	15
33	+	+	14	0
43	−	+	12	4
53	+	+	8	3
63	−	+	8	6
73	−	−	6	5
04	+	+	13	7
14	−	+	9	5
24	+	+	12	8
34	−	+	11	9
44	+	+	9	7
54	−	+	9	7
64	−	−	6	4
05			7	0
15	−	−	9	7
25	−	+	7	4
35	−	−	7	5
45	−	+	8	7
55	−	−	5	0
65	−	+	7	5
06	−	−	6	3
16	−	+	5	3
26	−	−	6	5
36	−	+	6	4
46	−	−	6	4
56	−	+	6	4

The two projections are shown in Figures 9 and 10 respectively. The broken contour line divides one of the equal intervals of potential indicated by the full lines. In each case the arrangement of the molecules at the centre and corners of the unit cell is shown clearly, and the projections of the two rows of carbon atoms in the molecule are well resolved. However, the projection obtained from the uncorrected data contains a number of small spurious maxima. The correction for secondary scattering removes much of this false detail and allows the positions of the hydrogen atoms to be seen clearly. For comparison a skeleton molecule is sketched with C–C distance 0·90 A. in projection, the C–H distance 1·05 A. and an angle of 109° between C–H bonds.

Although uncorrected intensities may allow an estimation of the coarser features of a structure, correction for secondary scattering is necessary for the detection of relatively light atoms or the accurate determination of atomic coordinates. With the aid of such corrections it is possible to take advantage of the fact that hydrogen has greater relative scattering power for electrons than for X-rays, and to use electron diffraction for the detection of hydrogen atoms in

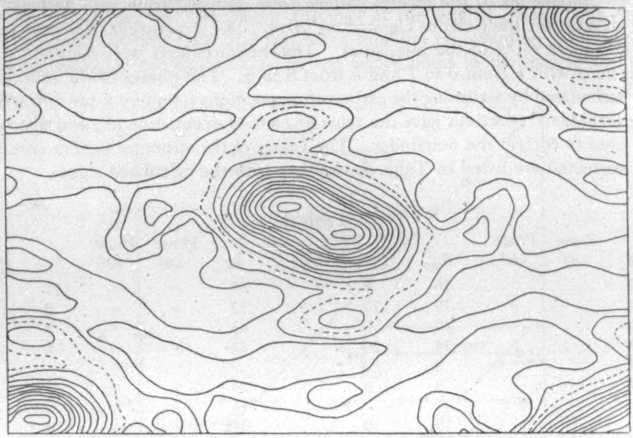


Figure 9. Basal-plane Fourier projection of dicetyl from observed intensities.

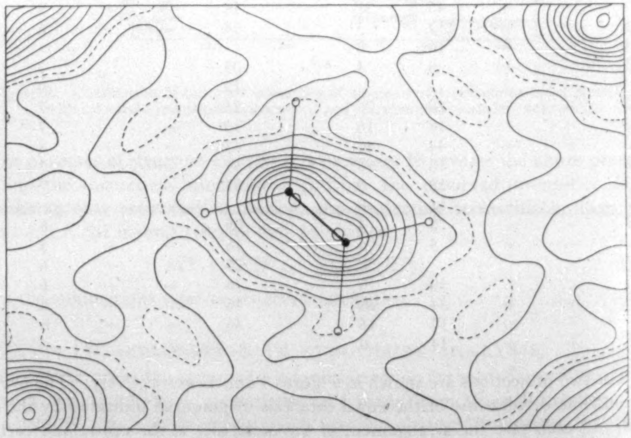


Figure 10. Basal-plane Fourier projection of dicetyl from intensities corrected for secondary scattering.

organic and other crystals. With more refined intensity measurements on more extensive patterns than those considered above the method could be used to investigate detail in quite complex structures.

REFERENCES

BUNN, C. W., 1939, *Trans. Faraday Soc.*, **35**, 482.
DARBYSHIRE, J. A., and COOPER, E. R., 1935, *Proc. Roy. Soc.* A, **152**, 104.
FINCH, G. I., and FORDHAM, S., 1937, *Chem. Industry*, **56**, 632.
FINCH, G. I., and SUN, C. H., 1936, *Trans. Faraday Soc.*, **32**, 852.
FINCH, G. I., and WILMAN, H., 1936, *Trans. Faraday Soc.*, **32**, 1539; 1937, *Ergebn. exakt. Naturw.*, **16**, 353.
KIRCHNER, P., 1932, *Ergebn. exakt. Naturw.*, **11**, 64.
RAETHER, H., 1932, *Z. Phys.*, **78**, 527.
TAYLOR-JONES, E., 1934, *Phil. Mag.*, **18**, 291.
VANSHSTEIN, B. K., and PINSKER, Z. G., 1949, *J. Phys. Chem., U.S.S.R.*, **23**, 1058; 1950, *Dokl. Akad. Nauk., S.S.S.R.*, **72**, 53.
WILMAN, H., 1940, *Proc. Phys. Soc.*, **52**, 325; 1950, *Nature, Lond.*, **165**, 321.

Nuclear Excitation by Electric Interaction with Charged Particles

BY R. HUBY AND H. C. NEWNS
Department of Theoretical Physics, Liverpool University

Communicated by H. Fröhlich; MS. received 7th October 1950 *and in final form 2nd February* 1951

ABSTRACT. Inelastic scattering of charged particles can take place through the electrostatic interaction between the particles and the nucleus. Expressions for the total cross section of the process have been derived using several methods and are found to be in close agreement. The angular distributions are less reliable. Questions for more practical interest are discussed and conditions given for the best observation of the effect. The total cross sections are not negligible and certain experimental results can be explained. Inelastic nuclear scattering has been extensively used hitherto to obtain the positions of nuclear energy levels. It is suggested that more information about nuclear states might be provided by experiments exploiting the electrostatic interaction.

§1. INTRODUCTION

IN the collision of a charged particle ('projectile') with a nucleus, energy may be exchanged between them through the interaction of their electrostatic fields. Alternatively, the inelastic scattering may occur through formation of an intermediate nucleus, and re-emission. The latter process, and all other competing reactions via the intermediate nucleus, are inhibited, however, if the projectile's energy is much below the Coulomb barrier, and so this condition is especially favourable to uncomplicated observation of the first-mentioned process, although it turns out that in this case the excitation energy has to be rather small (§4(i)).

The theory and best application of the electrostatic excitation of nuclei are here considered. The possibility of the process is well known and has been exploited experimentally, but a detailed theoretical appraisal of it has been lacking.* Weisskopf (1938) gave formulae for the total cross section, but without derivation, and confidence in his results may have been weakened both by an apparently contrary approach by Guth (1945) and by a statement as to the disagreement of the formulae with experiment (Risser, Lark-Horovitz and Smith 1940).

The total cross section is calculated in § 2. As rather crude approximations must be used, several different methods are taken (§ 2 (ii), (iii), (iv)) and their substantial agreement is demonstrated in § 2 (v). The results essentially reproduce Weisskopf's, apart from some rather significant numerical factors, and are not inconsistent with Guth's.

The angular distribution is discussed in § 3. The conclusions of practical application are drawn in § 4, where the order of magnitude of the expected cross sections is estimated, and shown to be not negligible. The optimum conditions for observation are investigated. It is demonstrated that the mechanism considered can account for a number of experimental observations of inelastic scattering which have been made, but not for all. Finally, it is pointed out that more complete measurements of such collisions might yield more kinds of information about the states of the nucleus involved than has been previously attempted.

§ 2. TOTAL CROSS SECTION

(i) *Multipole Development of Interaction*

Before detailing the methods used to obtain expressions for the total cross section, we obtain a convenient form for the interaction between the projectile and the nucleus.

The nucleus is treated as a collection of point charges. The interaction between the projectile of charge ze and the nucleus is given by $H' = ze\Sigma_i(e/r_i')$, where the summation is over the nuclear protons, and r_i' is the distance from the i'th proton to the projectile. If the projectile lies outside the nucleus, we may use the expansion in Legendre functions:

$$\frac{1}{r_i'} = \sum_{l=0}^{\infty} \frac{r_i^l}{R^{l+1}} P_l(\cos\theta_i').$$

(R, Θ, Φ) and (r_i, θ_i, ϕ_i) are the polar coordinates of the incident particle and the i'th proton respectively, and θ_i' is the angle between $\mathbf{R}$ and $\mathbf{r}_i$. Using the addition theorem for Legendre polynomials, viz.

$$P_l(\cos\theta_i') = \sum_{m=-l}^{l} \frac{2}{(2l+1)} \overline{P}_l^m(\cos\Theta)\overline{P}_l^m(\cos\theta_i)\exp\{im(\Phi-\phi_i)\},$$

where $\overline{P}_l^m$ denotes a normalized, associated Legendre function, we obtain

$$H' = ze^2 \sum_{l=0}^{\infty}\sum_{m=-l}^{l}\left(\frac{2}{2l+1}\right)^{1/2}\frac{\overline{P}_l^m(\cos\Theta)}{R^{l+1}}e^{im\Phi}Q_l^m, \qquad \ldots\ldots(1)$$

where
$$Q_l^m = \Sigma_i\left(\frac{2}{2l+1}\right)^{1/2} r_i^l\overline{P}_l^m(\cos\theta_i)\exp(-im\phi_i) \qquad \ldots\ldots(1\,a)$$

is an expression depending only on the coordinates of the nuclear protons, and is simply a nuclear multipole moment. Only configurations in which the projectile is outside the nucleus will be assumed to contribute to the electric excitation, and so (1) is the expression to be used for H'.

---

* Since the completion of this paper, Professor E. Guth has kindly sent us a manuscript of a study on somewhat similar lines by J. Mullin and himself, to be published in the *Physical Review*.

(ii) *The Impact Parameter Method*

The projectile is considered as a classical point charge, exerting a Coulomb field on the nucleus, whose constituent particles are dealt with wave mechanically. The path of the projectile is taken to be a straight line, at a normal distance p from the centre of the nucleus. The Schrödinger equation for the nucleus is $(H^0+H')\chi=-(\hbar/i)\partial\chi/\partial t$, where H^0 is the undisturbed Hamiltonian for the nucleus. We expand χ in the form

$$\chi=\Sigma_p c_p(t)\exp(-iW_p t/\hbar)\chi_p,$$

where the unperturbed wave function χ_p and energy W_p satisfy $H^0\chi_p=W_p\chi_p$. Then using first order perturbation theory, we find

$$c_n(\infty)=-\frac{i}{\hbar}\int_{-\infty}^{\infty}(n\,|\,H'\,|\,0)e^{i\omega t}\,dt,$$

where $\omega=(W_n-W_0)/\hbar$, and $(n\,|\,H'\,|\,0)=\int\chi_n{}^*H'\chi_0\,d\tau$. We now take the origin of coordinates at the centre of the nucleus and z axis parallel to the path of the projectile. Putting $t=Z/v$, where v is the velocity of the projectile, and using the expression for H' given by (1), we obtain

$$c_n(\infty)=-\frac{ize^2}{\hbar v}\sum_{l=1}^{\infty}\sum_{m=-l}^{l}\left(\frac{2}{2l+1}\right)^{1/2}e^{im\Phi}(n\,|\,Q_l{}^m\,|\,0)I_l{}^m,$$

where
$$I_l{}^m=\int_{-\infty}^{\infty}\bar{P}_l{}^m\frac{(\cos\theta)}{R^{l+1}}\exp(iZ/p_0)\,dZ, \qquad \ldots\ldots(2)$$

with
$$p_0=v/\omega=v\hbar/(W_n-W_0). \qquad \ldots\ldots(2\,a)$$

The element of cross section for excitation of the state n, due to paths lying within distances $p-(p+dp)$, is

$$\begin{aligned}d\sigma&=p\,dp\int_0^{2\pi}|\,c_n(\infty)\,|^2\,d\Phi\\&=4\pi p\,dp\,\frac{z^2e^4}{\hbar^2v^2}\sum_{l=1}^{\infty}\sum_{l'=1}^{\infty}\sum_{m=-l}^{l}\frac{(n\,|\,Q_l{}^m\,|\,0)(n\,|\,Q_{l'}{}^m\,|\,0)^*I_l{}^mI_{l'}{}^{m*}}{(2l+1)^{1/2}(2l'+1)^{1/2}}. \qquad \ldots\ldots(3)\end{aligned}$$

We now suppose that the initial state 0 and the final state n of the nucleus are characterized by quantum numbers J_{i}, M_{i} and J_{f}, M_{f} respectively. The nucleus in the initial state can have any of $(2J_{\mathrm{i}}+1)$ values of M_{i} and can go to any of $(2J_{\mathrm{f}}+1)$ values for M_{f}. We average over the initial magnetic quantum numbers M_{i} and sum over the final quantum numbers M_{f}. Then it can be shown by a group-theoretical method that

$$\frac{1}{(2J_{\mathrm{i}}+1)}\sum_{M_{\mathrm{i}},\,M_{\mathrm{f}}}(J_{\mathrm{f}}M_{\mathrm{f}}\,|\,Q_l{}^m\,|\,J_{\mathrm{i}}M_{\mathrm{i}})(J_{\mathrm{f}}M_{\mathrm{f}}\,|\,Q_{l'}{}^{m'}\,|\,J_{\mathrm{i}}M_{\mathrm{i}})^*=\frac{\delta_{ll'}\delta_{mm'}}{(2l+1)}|(f\,|\,Q_l\,|\,i)\,|^2 \quad \ldots(4)$$

where
$$|(f\,|\,Q_l\,|\,i)\,|^2=\sum_{M_{\mathrm{i}},\,M_{\mathrm{f}},\,\bar{m}}\frac{|(J_{\mathrm{f}}M_{\mathrm{f}}\,|\,Q_l{}^{\bar{m}}\,|\,J_{\mathrm{i}}M_{\mathrm{i}})|^2}{(2J_{\mathrm{i}}+1)}. \qquad \ldots\ldots(4\,a)$$

$|(f\,|\,Q_l\,|\,i)\,|^2$ is a number representing the total strength of the transition matrix elements of all the multipole moments $Q_l{}^{\bar{m}}$, of order l, leading from any state in the initial degenerate level i to the final degenerate level f. Using (4) together with (3), we obtain for the averaged and summed element of cross section

$$d\sigma=4\pi p\,dp\,\frac{z^2e^4}{\hbar^2v^2}\sum_{l=1}^{\infty}\sum_{m=-l}^{l}\frac{|(f\,|\,Q_l\,|\,i)\,|^2\,|\,I_l{}^m\,|^2}{(2l+1)^2}. \qquad \ldots\ldots(5)$$

Evaluation of the integral I_l^m, defined by (2). We may substitute $Z=p\cot\Theta$, and $R=p\operatorname{cosec}\Theta$, and integrate over Θ from 0 to π. The integration is difficult, but can be readily performed if the factor $\exp(iZ/p_0)=\exp(ip/p_0\cot\Theta)$ is replaced by unity. This is clearly justified provided $p\ll p_0$, while when $p>p_0$ the integral is negligible, for the disturbance due to such a distantly passing projectile is so slowly varying that the nucleus responds adiabatically without becoming excited. Making this simplification

$$I_l^m=\frac{1}{p^l}\int_0^\pi \bar{P}_l^m(\cos\Theta)\sin^{l-1}\Theta\,d\Theta=\begin{cases}0 \text{ for } m\neq l,\\ \dfrac{\pi^{1/4}}{p^l}\dfrac{(l+\frac{1}{2})^{1/2}}{l}\left[\dfrac{l!}{(l-\frac{1}{2})!}\right]^{1/2} \text{ for } m=l.\end{cases} \qquad \ldots.(6)$$

Thus only the multipole moments $P_l^l(\cos\Theta)e^{il\Phi}$ make any contribution.

Total cross section. To obtain the total cross section, we must now integrate (5) over the range of impact parameters, using (6) for the integral I_l^m. We have seen that there is an effective upper cut-off at $p=p_0$ (equation (2 *a*)), so we integrate from a lower limit p_1 up to p_0. The possible values to be assigned to p_1 will be discussed in (v). The result is

$$\sigma=\sum_{l=1}^{\infty}\sigma_l=\frac{z^2e^4}{\hbar^2v^2}\pi^{3/2}\left\{\frac{8}{3\pi^{1/2}}\ln(p_0/p_1)\,|(f|Q_1|i)|^2\right.$$
$$\left.+\sum_{l=2}^{\infty}\frac{|(f|Q_l|i)|^2(l-2)!}{l(l+\frac{1}{2})!}\left(\frac{1}{p_1^{2l-2}}-\frac{1}{p_0^{2l-2}}\right)\right\}. \qquad \ldots\ldots(7)$$

This is the expression for the total cross section σ for the process, which is thus the sum of contributions σ_l from the individual multipoles.

The formula applies only if $p_0\gg p_1$, in which case the term $1/p_0^{2l-2}$ can be neglected; otherwise the contribution of each multipole $(f|Q_l|i)$ will be smaller by a factor of the order of $\exp(-2p_1/p_0)$.

(iii) *The Born Approximation*

An expression for the total cross section can also be obtained using Born's approximation. The differential cross section for excitation of nuclear state n using Born's approximation is given by

$$\sigma(\theta)=\frac{k_nm^2}{k_04\pi^2\hbar^4}\left|\int\exp\{i(\mathbf{k}_0-\mathbf{k}_n)\mathbf{R}\}\chi_n{}^*H'\chi_0\,d\tau\,d\mathbf{R}\right|^2,$$

where m is the reduced mass of the system, $k_0=mv_0/\hbar$ and $k_n=mv_n/\hbar$ are the incoming and outgoing wave numbers respectively, and $d\tau$ represents a volume element of the internal coordinates of the nucleus. Using expressions (1) for H', and integrating over the coordinates of the nucleus, this becomes

$$\sigma(\theta)=\frac{k_n}{k_0}\frac{z^2m^2e^4}{4\pi^2\hbar^4}\left|\sum_{l=1}^{\infty}\sum_{m=-l}\left(\frac{2}{2l+1}\right)^{1/2}\int\exp\{i(\mathbf{k}_0-\mathbf{k}_n)\mathbf{R}\}\right.$$
$$\left.\times\frac{\bar{P}_l^m(\cos\Theta)e^{im\Phi}}{R^{l+1}}(n|Q_l^m|0)\,d\mathbf{R}\right|^2. \qquad \ldots\ldots(8)$$

Expanding,

$$\exp i(\mathbf{k}_0-\mathbf{k}_n)\mathbf{R}=\exp(i\mathbf{k}\mathbf{R})=\sum_{l=0}^{\infty}\sum_{m=-l}^{l}(2\pi)^{1/2}i^l\frac{J_{l+\frac{1}{2}}(kR)}{(kR)^{1/2}}\bar{P}_l^m(\cos\Theta)\bar{P}_l^m(\cos\theta')\,e^{im(\varphi'+\Phi)},$$

where (θ', ϕ') are the angles defining the direction of $\mathbf{k}=\mathbf{k}_0-\mathbf{k}_n$. Using this relation in (8), the integrations over the projectile coordinates can be carried out and lead to the result

$$\sigma(\theta)=\frac{k_n}{k_0}\frac{4\pi z^2e^4m^2}{\hbar^4}\left|\sum_{l=1}^{\infty}\sum_{m=-l}^{l}\frac{i^lk^{l-2}}{(2l+1)^{1/2}}\frac{J_{l-\frac{1}{2}}(ka)}{(ka)^{l-\frac{1}{2}}}\overline{P}_l^m(\cos\theta')e^{im\varphi'}(n\,|Q_l^m\,|0)\right|^2.$$

In the integration over R we have cut off at a lower limit a which corresponds to the lower impact parameter p_1 of the previous method. Discussion of possible values for a will be given in (v). If we now average over the initial magnetic quantum numbers M_i of the nucleus, and sum over the final ones M_f as before, then after using (4) and summing over m we obtain

$$\sigma(\theta)=\sum_{l=1}^{\infty}\sigma_l(\theta)=\frac{k_n}{k_0}\frac{2\pi z^2e^4m^2}{\hbar^4}\sum_{l=1}^{\infty}\frac{1}{2l+1}\left[\frac{J_{l-\frac{1}{2}}(ka)}{(ka)^{l-\frac{1}{2}}}k^{l-2}\right]^2|(f\,|Q_l|\,i)\,|^2. \qquad \ldots\ldots(9)$$

The scattering angle θ, i.e. the angle between $\mathbf{k}_0$ and $\mathbf{k}_n$, is contained in this expression in the quantity k, for $k^2=k_0^2+k_n^2-2k_0k_n\cos\theta$. To obtain the total cross section we have to integrate over the scattering angle $\sigma=2\pi\int_0^{\pi}\sigma(\theta)\sin\theta\,d\theta$.

Essentially the integral we have to evaluate is $\int_0^{\pi}\left[\frac{J_{l-\frac{1}{2}}(ka)}{(ka)^{l-\frac{1}{2}}}k^{l-2}\right]^2\sin\theta\,d\theta$.

By a suitable substitution, this becomes proportional to

$$\int_A^B\left[\frac{J_{l-\frac{1}{2}}(u)}{u}\right]^2du, \qquad \ldots\ldots(10)$$

with $A=(k_0-k_n)a$, $B=(k_0+k_n)a$. In general this integral is tedious to evaluate, but in the circumstances of interest, A will often be small and B large (see (v)), so that (10) may be replaced by

$$\int_0^{\infty}\left[\frac{J_{l-\frac{1}{2}}(u)}{u}\right]^2du=\frac{1}{\pi l(l-1)}.$$

This is only valid for l greater than or equal to 2, since in the dipole case $(l=1)$ (10) diverges when A tends to 0. However, (10) can be evaluated by elementary means for $l=1$. The result for the total cross section in this approximation is

$$\sigma=\frac{4\pi z^2e^4m^2}{k_0^2\hbar^4}\left\{\tfrac{2}{3}\ln\left(\frac{1\cdot 3}{A}\right)|(f\,|Q_1|\,i)\,|^2+\sum_{l=2}^{\infty}\frac{1}{a^{2l-2}}\frac{|(f\,|Q_l|\,i)\,|^2}{l(l-1)(2l+1)}\right\}. \qquad \ldots\ldots(11)$$

When $k_0a\ll 1$ (while $a\geqslant$ the radius of the nucleus) the effect of cutting off the radial integral at $R=a$ is not appreciably different from that of continuing the electric interaction right through the nucleus to the origin. This case will therefore be referred to as the 'true Born approximation', for which the differential cross section (9) becomes

$$\sigma(\theta)=\frac{k_n}{k_0}\frac{4z^2e^4m^2}{\hbar^4}\sum_{l=1}^{\infty}\frac{1}{(2l+1)}\left[\frac{2^{l-1}(l-1)!}{(2l-1)!}\right]^2k^{2l-4}\,|(f\,|Q_l|\,i)\,|^2. \qquad \ldots\ldots(12)$$

The total cross section is obtained quite simply by integration over θ:

$$\sigma=\frac{8\pi z^2e^4m^2}{k_0^2\hbar^4}\left\{\tfrac{1}{3}\ln\left(\frac{k_0+k_n}{k_0-k_n}\right)\cdot|(f\,|Q_1|\,i)\,|^2\right.$$
$$\left.+\sum_{l=2}^{\infty}\left[\frac{2^l l!}{(2l+1)!}\right]^2\frac{(2l+1)}{2(l-1)}[(k_0+k_n)^{2l-2}-(k_0-k_n)^{2l-2}]\,|(f\,|Q_l|\,i)\,|^2\right\}. \qquad \ldots\ldots(13)$$

(iv) *Method of Distorted Waves*

An improvement over the Born approximation, particularly important for projectile energies below the Coulomb barrier, is effected by taking the Rutherford scattering into account exactly, while still treating the multipole part of the electrostatic interaction ($l>0$) as a perturbation. The differential cross section for excitation of the nuclear state n is then to first order

$$\sigma(\theta)=\frac{k_n}{k_0}\frac{m^2}{4\pi^2\hbar^4}\left|\int\tilde{\psi}^*(\mathbf{k}_n,\mathbf{R})\psi(\mathbf{k}_0,\mathbf{R})\chi_n{}^*H'\chi_0\,d\tau\,d\mathbf{R}\right|^2. \qquad \ldots\ldots(14)$$

Here ψ $(\mathbf{k},\mathbf{R})$ is the exact wave function for a particle of mass m and charge z in the field of a charge Z (the nuclear charge), when ψ has the asymptotic form of a plane wave $\exp\{i(\mathbf{k}.\mathbf{R})\}$, plus an outgoing radial wave. $\tilde{\psi}(\mathbf{k},\mathbf{R})$ differs only in that it tends asymptotically to a plane wave plus an incoming radial wave, and it is in fact $\psi^*(-\mathbf{k},\mathbf{R})$. We have

$$\psi(\mathbf{k},\mathbf{R})=\left[\frac{2\pi\alpha}{e^{2\pi\alpha}-1}\right]^{1/2}\exp\{i(\mathbf{k}.\mathbf{R})\}.F(-i\alpha\,;1\,;i\{kR-(\mathbf{k}.\mathbf{R})\}), \qquad \ldots\ldots(15)$$

where

$$\alpha=zZe^2m/k\hbar^2=zZe^2/\hbar v. \qquad \ldots\ldots(15\,a)$$

It has not been found possible to evaluate $\sigma(\theta)$ from (14) using H' as given by (1). However, the contributions from the dipole terms alone in H' ($l=1$) have been calculated, the integral being taken over all space, i.e. without cut-off as in (iii). The procedure employed to obtain the results (18) and (19) below, due originally to Professor E. Guth, was passed on to us by Professor R. Peierls. Substituting the dipole terms in (14), averaging the cross section over initial quantum numbers M_{i}, and summing over final quantum numbers M_{f}, we find with the help of (4):

$$\sigma_1(\theta)=\frac{k_n}{k_0}\frac{m^2z^2e^4}{4\pi^2\hbar^4}\frac{|(f\,|\,Q_1\,|\,i)|^2}{3}\left|\left(\mathrm{grad}\,\frac{1}{R}\right)_{n,0}\right|^2, \qquad \ldots\ldots(16)$$

where

$$\left(\mathrm{grad}\,\frac{1}{R}\right)_{n,0}=\int\tilde{\psi}^*(\mathbf{k}_n,\mathbf{R})\,\mathrm{grad}\,\frac{1}{R}\,\psi(\mathbf{k}_0,\mathbf{R})\,d\mathbf{R}.$$

The last quantity can be transformed, by appeal to the differential equation satisfied by the ψ functions, to:

$$\left(\mathrm{grad}\,\frac{1}{R}\right)_{n,0}=\frac{\hbar^2(k_0{}^2-k_n{}^2)}{2mzZe^2}\int\tilde{\psi}^*(\mathbf{k}_n,\mathbf{R})\,\mathrm{grad}\,\psi(\mathbf{k}_0,\mathbf{R})\,d\mathbf{R}. \qquad \ldots\ldots(17)$$

This integral is obtainable from calculations on bremsstrahlung given in Sommerfeld (1945) with the result

$$\left|\left(\mathrm{grad}\,\frac{1}{R}\right)_{n,0}\right|^2=-\frac{32\pi^4}{(k_0-k_n)^2}\frac{\exp(2\pi\alpha_0)}{\{\exp(2\pi\alpha_0)-1\}\{\exp(2\pi\alpha_n)-1\}}\frac{d}{dx}\left[x\frac{d}{dx}\,|\,F(x)\,|^2\right], \qquad \ldots\ldots(18)$$

where $x=-\{4k_0k_n/(k_0-k_n)^2\}\sin^2\frac{1}{2}\theta$ and $F(x)=F(-i\alpha_0,-i\alpha_n\,;1\,;x)$. α_0 and α_n are given by substituting k_0 and k_n respectively for k in (15 *a*). To obtain the total cross section we shall require the integral of this expression over θ, which yields:

$$2\pi\int_0^\pi\left|\left(\mathrm{grad}\,\frac{1}{R}\right)_{n,0}\right|^2\sin\theta\,d\theta$$

$$=-\frac{128\pi^5}{(k_0-k_n)^2}\frac{\exp(2\pi\alpha_0)}{\{\exp(2\pi\alpha_0)-1\}\{\exp(2\pi\alpha_n)-1\}}\frac{d}{dx_0}\,|\,F(x_0)\,|^2, \qquad \ldots\ldots(19)$$

with $x_0 = -4k_0k_n/(k_0-k_n)^2$. It may be noted that the expression for the forward scattering given by (18) is simple:

$$\sigma_1(0) = \frac{k_n}{k_0}\frac{4}{3}\frac{m^2z^2e^4}{\hbar^4}\exp(2\pi\alpha_0)\frac{2\pi\alpha_0}{\{\exp(2\pi\alpha_0)-1\}}\frac{2\pi\alpha_n}{\{\exp(2\pi\alpha_n)-1\}}|(f|Q_1|i)|^2. \qquad \ldots\ldots(20)$$

Expressions (17) and (18) can be reduced considerably in a case which will be of interest in (v) below, viz. when

$$2\pi(\alpha_n-\alpha_0)\ll 1, \qquad \ldots\ldots(21) \qquad \text{and} \qquad k_0-k_n\ll k_0. \qquad \ldots\ldots(22)$$

Under these conditions, and for angles θ sufficiently large that $(\alpha_n-\alpha_0)/2\sin\theta/2\ll 1$, $F(x)$ may be transformed into hypergeometric functions which can be expanded in powers of $1/x$ with the approximate result:

$$\frac{d}{dx}|F(x)|^2 = \frac{1}{x\pi^2}\frac{\{\exp(2\pi\alpha_0)-1\}\{\exp(2\pi\alpha_n)-1\}}{\exp(2\pi\alpha_0)}[\tfrac{1}{2}\ln(-x)+\Psi(0)-R\Psi(i\alpha_n)], \qquad \ldots\ldots(23)$$

where Ψ is the logarithmic derivative of the factorial function. Substitution of this in (18) and (19) and thence into (14), yields for the differential cross section, at the angles just indicated above,

$$\sigma_1(\theta) = \frac{1}{3}\frac{m^2e^4}{\hbar^4k_0^2}\frac{|(f|Q_1|i)|^2}{\sin^2\frac{1}{2}\theta}, \qquad \ldots\ldots(24)$$

and for the total cross section

$$\sigma_1 = \frac{8}{3}\frac{\pi z^2e^4m^2}{\hbar^4k_0^2}|(f|Q_1|i)|^2\left[\ln\frac{2mv^2}{(W_n-W_0)}+\Psi(0)-R\Psi\left(\frac{izZe^2}{v_n\hbar}\right)\right].^{*} \qquad \ldots\ldots(25)$$

A different approximation can be made in (17) and (18) if the velocities of incoming and outgoing particles are so small that α_0, α_n and $(\alpha_n-\alpha_0)$ are large. The approximation to $F(x)$ in this case is also given by Sommerfeld (1945). The governing factor in the cross section then turns out to be $\exp\{-2\pi(\alpha_n-\alpha_0)\}$, as was pointed out by Guth (1945).

(v) *Validity and Comparison of Methods*

The conditions of validity of the three methods developed will be investigated, and it will be shown that under corresponding conditions they agree, up to a numerical factor. The impact parameter method presents the most perspicuous picture, but is theoretically the least well founded, and so calls for the corroboration of the other methods.

In the integration over impact parameters, a natural upper cut-off was provided at p_0 (equation (2 *a*)), and the lower cut-off p_1 has yet to be determined. Three factors interfere with passage of the projectile near the nucleus:

(*a*) *The Coulomb repulsion.* This curves the track into a hyperbola, and in particular imposes a distance of closest approach

$$r_c = 2zZe^2/mv_n^2. \qquad \ldots\ldots(26)$$

(The final velocity v_n is here used rather than the initial velocity v_0, as giving the larger value of r_c.) This may be very crudely represented by supposing that

---

[*] This formula has a remarkable but, it seems, largely fortuitous affinity with the well-known formula of Bloch (1933) for the energy loss of a fast particle due to inelastic scattering by atoms, calculated by a modified impact parameter method.

all tracks are undeflected up to a distance $\gamma_A r_c$, with γ_A some constant of order unity, and that there are no closer tracks whatever. Then p_1 is

$$p_A = \gamma_A r_c. \qquad \ldots\ldots(27)$$

(*b*) *Wave nature of projectile.* The classical model of the projectile breaks down at distances less than approximately $1/k_n$. This may be taken to limit the impact parameter at

$$p_B = \gamma_B / k_n = \gamma_B \hbar / m v_n, \qquad \ldots\ldots(28)$$

with γ_B of order unity. The quantities k_n and v_n are here used for the same reason as v_n in (*a*).

(*c*) *Size of nucleus.* We consider only excitation while the projectile is outside the nucleus. This may be supposed to impose a lower limit

$$p_C = \text{radius of nucleus, } r_n. \qquad \ldots\ldots(29)$$

In any experiment, the operative value of p_1 must be the largest of p_A, p_B, p_C. At the lowest energies p_A will be limiting, and at the highest p_C, with p_B either at intermediate energies or completely excluded. It is desirable to choose the numbers γ_A and γ_B so that eventually the impact parameter results shall agree as closely as possible with those of the other methods. However, it is found that no choice can effect this with perfect consistency. The rather arbitrary values which we shall adopt for calculation are $\gamma_A = \frac{1}{2}$, $\gamma_B = 1$.

The upper impact parameter p_0 must always be much greater than all three of p_A, p_B, p_C (see end of (ii)). It may be noted that one of these requirements, $p_0 \gg p_B$, can be seen from (2 *a*) and (28) to mean approximately that

$$W_n - W_0 \ll \tfrac{1}{2} m v_n^2, \qquad \ldots\ldots(30)$$

which is an obvious precondition for the impact-parameter approximation.

Before considering the other methods, the following relations may be noted between the parameters k_0, k_n, α_0, α_n (see (15 *a*)) important in those methods, on the one hand, and some of the limiting impact parameters just discussed, on the other hand:

$$p_0 = \hbar v / (W_n - W_0) = 1/(k_0 - k_n) = \alpha_n / (\alpha_n - \alpha_0) k_n, \qquad \ldots\ldots(31)$$

which is valid provided (30) holds; and

$$r_c = 2\alpha_n / k_n, \qquad \ldots\ldots(32)$$

which follows from (15 *a*) and (26).

We turn to the Born approximation. The 'true' form of this (equation (13)), i.e. without cut-off, should apply under the conditions which cause the impact parameter method to be limited by wave effects alone, i.e. $p_1 = p_B$. Substituting then (28) in the impact-parameter formula (7), and allowing for (30) in the true Born approximation formula (13), we find that the cross sections for the two methods are identical, except in the following respects: (*a*) dipolar term: the argument of the logarithm is $2\gamma_B$ times as large in the Born result as the impact parameter; (*b*) multipole terms ($l > 1$): the ratio of the lth term in the impact parameter case to that in the Born case is $(2l)!\,\gamma_B^{2l-2}/2^{2l-1} l^2$.

Under conditions which make the impact parameter p_1 greater than p_B, it seems reasonable to apply the Born approximation with cut-off of the matrix element integration at a minimum radius a equal to p_1. The Born formula is then

(11), provided $(k_0-k_n)p_1 \ll 1$, and $(k_0+k_n)p_1 \gg 1$ (see below (10)), which, as follows from (28) and (31), are roughly equivalent respectively to the impact-parameter requirements $p_1 \ll p_0$ and $p_1 \gg p_B$, and are therefore valid in this case. The Born result (11) is now identical with the impact parameter (7), except in the following respects: (*a*) dipolar term: the argument of the logarithm is 1·3 times as great in the Born result as the impact parameter; (*b*) multipole terms ($l>1$): the ratio of the lth term in the impact parameter case to that in the Born case is $\pi^{1/2}(l-1)!/2(l-\frac{1}{2})!$. This is 2/3 when $l=2$, 8/15 when $l=3$, 48/105 when $l=4$. Thus under all conditions in which the impact parameter method may be used, it gives the same results as our adaptation of the Born approximation, to an order of magnitude.

It remains to be considered whether the means provided in these methods for taking into account the Coulomb repulsion is valid. A check on the Coulomb repulsion is provided by the method of distorted waves, which deals with this effect exactly, though only dipole excitation has been calculated, with neglect of the finite size of the nucleus. This method yields (25), provided (21) and (22) are satisfied. The condition (21) is shown by (27), (31) and (32) to be equivalent, apart from a numerical factor, to the impact-parameter requirement that p_A shall always be very much less than p_0, while (22) is likewise almost equivalent to (30), or $p_B \ll p_0$. There are two extreme cases:

(*a*) *Coulomb repulsion negligible*, i.e. $p_B \gg p_A$, or, according to (27), (28) and (32), $\alpha_n \ll 1$. Then with the aid of (15 *a*) the distorted waves result (25) becomes identical with the first term of the true Born result (13).

(*b*) *Coulomb repulsion determinant*, i.e. $p_A \gg p_B$, or $\alpha_n \gg 1$. Then (25) becomes the same as the first term of the impact parameter result (7), if there p_1 is put equal to p_A, except that the argument of the logarithm is $2{\cdot}24\gamma_A$ times greater in (25) than in (7).

Thus the exact treatment of the Coulomb repulsion agrees very closely with the approximate.

The case of failure of the approximations may be noted. At very low velocities with $p_1=p_A$, the impact-parameter method may fail because $p_0<p_A$, and the cross section is then reduced by a factor of the order of $\exp(-2p_A/p_0)$ (see end of (ii)). This condition means $\alpha_n-\alpha_0 \gg 1$, and then, as pointed out at the end of (iv), the distorted wave result (25) fails equally, and a factor

$$\exp\{-2\pi(\alpha_n-\alpha_0)\}=\exp(-\pi p_A/\gamma_A p_0)$$

appears in the cross section. Although the exponential factors for the two methods are different, the general effect is similar.

Finally our results may be compared with the formulae given by Weisskopf (1938). He gives the cross section only for multipoles higher than dipole, in two cases corresponding nearly to those in which we take $p_1=p_A$ or p_C. In both cases, if we assume (22), his formulae become the same as ours, except for numerical factors. In the case $p_1=p_A$, if we put $\gamma_A=\frac{1}{2}$, the agreement is quite close. Thus the ratio of Weisskopf's cross section to the impact-parameter value is 12·6, 5·3, 1·9, 0·67, for $l=2$, 3, 4, 5 respectively.

In the case $p_1=p_C$, the discrepancy is serious, the ratio of Weisskopf's cross section to the impact-parameter value being 75, 106, 147, 193 for $l=2$, 3, 4, 5 respectively.

§ 3. ANGULAR DISTRIBUTION OF SCATTERED PARTICLES

The differential cross sections obtained in the last section will now be considered in more detail and the angular distribution investigated. Here the results of the Born approximation must be used, although in the single case of a dipole ($l=1$) transition there is also available the distorted waves method. It will be seen that in this case there are serious discrepancies between the results of the two methods which make the angular distributions obtained very unreliable.

We first consider the true Born approximation where the cut-off radius a tends to zero. The differential cross section in this case is given by equation (12). This gives

$$\sigma_l(\theta) \propto k^{2l-4} \propto [(k_0-k_n)^2+4k_0k_n\sin^2\tfrac{1}{2}\theta]^{l-2}. \qquad \ldots\ldots(33)$$

These angular distributions are all smooth curves giving for a dipole ($l=1$) transition the form of an ellipse with a maximum in the forward direction in the polar diagram. The quadrupole ($l=2$) transition is spherically symmetrical and octopole ($l=3$) transition is similar to the dipole case except that now the maximum is in a backward direction. For $l>3$ the scattering continues to be mainly in the backward direction.

It will be shown in the discussion of the values for the lower cut-off parameters p_1 and a that the middle range ($p_1=p_{\mathrm{B}}$, $a\to 0$) does not apply and we must cut off either at the distance of closest approach or at the radius of the nucleus.

From equation (9) the differential cross section when cutting off at some lower parameter a is

$$\sigma_l(\theta) \propto [k^{l-2}\{J_{l-\frac{1}{2}}(ka)\}/(ka)^{l-\frac{1}{2}}]^2, \qquad \ldots\ldots(34)$$

which is simply proportional to (33) multiplied by the factor

$$[\{J_{l-\frac{1}{2}}(ka)\}/(ka)^{l-\frac{1}{2}}]^2. \qquad \ldots\ldots(35)$$

This function has a maximum at the origin and oscillates as the argument increases, but with ever decreasing amplitude. Consequently the backward scattering as given by (33) is reduced by a large factor in comparison with the forward scattering. This causes enormous forward maxima to appear in the distributions for dipole and quadrupole transitions. For higher multipole transitions, (33) increases faster than the multiplying factor (35) decreases. So a forward minimum is obtained which rises to a maximum at a certain angle. As the order of the multipole increases this maximum shifts further towards the backward direction.

We will now consider the results obtained using the distorted waves method for a dipole transition.

The scattering in the forward direction can be obtained from equation (20). The factors involving α_0 and α_n constitute the deviation from the Born formula and show a considerable reduction in the cross section, approximately equal, if α_0 and α_n are much greater than unity, to the diminution in the probability that the outgoing projectile is at the origin due to the Coulomb repulsion (factor $2\pi\alpha_n/\{\exp(2\pi\alpha_n)-1\}$).

For large angles it is permissible to use equation (24) provided equations (21) and (22) hold and that $(\alpha_n-\alpha_0)/2\sin\frac{1}{2}\theta \ll 1$. In this case, and provided $(k_0-k_n)/2k_n\sin\frac{1}{2}\theta \ll 1$, the dipole terms of the distorted wave (24) and true Born approximation (12) results become identical.

The distorted waves method, however, takes into account the effect of the Coulomb repulsion of the nucleus exactly, whereas this is attempted in the modified Born approximation by cutting off at the distance of closest approach. This procedure however introduces the factor (35) which causes a large diminution in the backward scattering as given by (33) and fails to show the forward diminution (20). This shows that the method by which we try to take into account the Coulomb repulsion in the Born approximation is extremely crude and gives rise to misleading results.

It is remarkable, however, that the agreement between the total cross sections obtained by the three methods is, as we have already seen, extremely close, and it seems justified to assume that the expressions obtained for the total cross sections are reasonably accurate.

We now consider the case of very high energy particles. This means that we must cut off at the radius of the nucleus in the Born approximation. As the Coulomb repulsion is negligible at high energy, use of the distorted waves $\psi(\mathbf{k}, \mathbf{R})$ would offer no improvement. The distribution (34) is now the appropriate one, but at these energies the effect would probably be masked by large nuclear scattering.

This mechanism is most important when the energies of the projectiles are below the nuclear barrier but, as we have seen, it has not been possible to obtain reliable angular distributions in this region.

§ 4. APPLICATIONS

We shall consider applications to typical experiments, basing the discussion mainly on the impact parameter picture.

(i) *Validity of Formulae*

Different formulae are obtained, according as the minimum impact parameter p_1 is equal to p_A, p_B or p_C (equations (27), (28), (29)). The case $p_1 = p_B$ does not arise if

$$r_n > \gamma_B^2 \hbar^2 / 2\gamma_A zZme^2. \qquad \ldots\ldots(36)$$

We assume

$$r_n = A^{1/3} r_0, \qquad \ldots\ldots(37)$$

with $r_0 = 1\cdot5 \times 10^{-13}$ cm.

As stated in § 2 (v), we shall take $\gamma_A = \frac{1}{2}$, $\gamma_B = 1$. The substitution of numerical values in (36) shows that the case $p_1 = p_B$ is excluded for proton projectiles if $Z > 7$, for deuterons if $Z > 4$ and for α-particles with all target nuclei. We shall generally assume this to be the case. Then p_1 is the larger of p_A and p_C, the former being the case for projectile energies roughly below the barrier height. The cross section is given by (7) with (27) or (29). Especially important is the question whether the electric excitation can still take place at incident energies well below the Coulomb barrier, so that reactions involving a compound state are inhibited by a factor of the order of $\exp\{-\pi(\alpha_0 + \alpha_n)\}$ (cf. equation (15)), while the incident energy is at the same time sufficiently high that the cross section for electric excitation is not seriously reduced by the factor $\exp\{-2\pi(\alpha_n - \alpha_0)\}$, (see end of § 2 (iv)). This is on condition that the excitation energy $(W_n - W_0)$ is sufficiently small. In fact, we require $\frac{1}{2}mv_0^2$ to be very much less than zZe^2/r_n, together with equation (21), and these simultaneous conditions are found with the help of (37) and (15 *a*) to be equivalent to

$$W_n - W_0 \ll \frac{\hbar e}{\pi}\left(\frac{2zZ}{mr_n^3}\right)^{1/2} = \left(\frac{Z}{m}\right)^{1/2} \times 1\cdot3 \text{ Mev.}, \qquad \ldots\ldots(38)$$

where m is expressed in units of the proton mass. Thus electric excitation unaccompanied by formation of a compound nucleus can only occur for rather low excitation energies. The minimum incident energy then compatible with (21) is

$$E_{\min} = \{(W_n - W_0)^2 z^2 Z^2 m\}^{1/3} \times 0{\cdot}63 \text{ MeV}. \qquad \ldots\ldots(39)$$

where W_n and W_0 are in MeV., and m is in proton masses.

It may be noted that the conditions just discussed take care of the less stringent conditions which must always be satisfied, viz. $p_0 > p_A$ and $p_0 > p_C$.

To take two typical examples, excitation of the 1·38 MeV. level of magnesium by a proton projectile does not satisfy (38), whereas the excitation of the 0·34 MeV. level of indium, also by a proton, does. In the latter case, the minimum incident energy as given by (39) would be about 4 MeV., which leaves a good range of energy available below the barrier height of about 10 MeV.

(ii) *Multipole Moments* $(f|Q_l|i)$

Multiple moments $(f|Q_l|i)$ are defined by equations (1 *a*) and (4 *a*). The number of different l's contributing to the cross section is restricted by the selection rules (*a*) $|J_f - J_i| \leqslant l \leqslant J_f + J_i$ and (*b*) l is even or odd according as the parities of the initial and final states are the same or different. The functions $Q_l^{\overline{m}}$ have been so defined (1 *a*) that the sum of $|Q_l^{\overline{m}}|^2$ over all values of $\overline{m}$, for given l, and averaged over all directions of the vectors $\mathbf{r}_i$, is $\Sigma_i r_i^{2l}$. Hence in particular $|(f|Q_1|i)|$ (see equation (4)) is equal to the dipole moment as usually defined, viz. $\{\Sigma_{M_f}|(J_f, M_f|\Sigma_i \mathbf{r}_i|J_i, M_i)|^2\}^{1/2}$. As an order of magnitude, it seems reasonable to take, for any non-vanishing matrix element in a transition to a low excited state, by other than dipole interaction,

$$|(f|Q_l|i)| = r_n^{\,l}. \qquad \ldots\ldots(40)$$

Dipole matrix elements are known to be much smaller than this formula would predict, by a factor of the order one-tenth to one-hundredth.

(iii) *Optimum Cross Section*

We shall here mostly neglect the dipole cross section because of the smallness of the dipole matrix element.

From equation (7) and the discussion of p_1 in § 4 (i), it follows that the cross section becomes a maximum, as function of projectile energy, when $p_1 = p_A = p_C$, that is, for an energy in the region of the barrier height, or precisely for E equal to γ_A times barrier height. Then (7) with (40) yields

$$\sigma_l = \frac{zme^2}{\gamma_A \hbar^2} r_0^{\,3} \frac{(l-2)!}{l(l+\frac{1}{2})!} \pi^{3/2} = zm \times 2{\cdot}0 \times 10^{-27} \text{ cm}^2 \quad \text{for } l = 2; \qquad \ldots\ldots(41)$$
$$= zm \times 0{\cdot}37 \times 10^{-27} \text{ cm}^2 \quad \text{for } l = 3$$

where m is in proton masses. Thus the cross section at optimum energy is approximately independent of the position of the target nucleus in the periodic table, but depends on the nature of the projectile. The heavier, more highly charged the projectile the better, an α-particle giving eight times as large an optimum cross section as a proton.

In the measurement of inelastic scattering, the effect may be masked by elastically scattered particles. The measurability may be gauged by the ratio Λ of the average inelastic differential cross section $\sigma/4\pi$ to the Rutherford cross section at, say, 90°. At the energy which makes the inelastic cross section a maximum (equation (41)), Λ is proportional to $zm/Z^{2/3}$. Thus the dependence of Λ on the nature of the projectile is the same as that of σ but Λ decreases as the target becomes heavier. For $A=24$, at the energy indicated, Λ is only about 0·1% for protons, and 1% for α-particles, assuming quadrupole excitation. At higher energies, Λ should be larger, as the Rutherford scattering should fall off more rapidly than the inelastic. As a rough estimate of the dipole cross section, at the energy for which the higher multipoles are given by (41), we may suggest $2m \times 5 \times 10^{-28}$ to $2m \times 5 \times 10^{-30}$ cm².

(iv) *Comparison with Experiment*

Few experiments of the type here considered have hitherto produced any information more detailed than the excitation energies. Excitation of aluminium and magnesium by protons and deuterons (Bush and Fulbright 1948, Rhoderick 1949, Greenlees, Kempton and Rhoderick 1949, Holt and Young 1949) gives cross sections seemingly much too large to be accounted for by the Coulomb interaction, in view of the maxima predicted by (41). Rhoderick (1949) points out that the excitation function for magnesium by protons indicates that the reaction necessitates penetration of the nucleus by the projectile. The conclusion would be that the process is a normal nuclear reaction, proceeding via a compound state. However, the angular distribution for excitation of magnesium by deuterons, obtained by Holt and Young (1949) shows a strong forward maximum, which suggests a one-stage rather than a two-stage process.

Holt and Young find excitation of a level at 0·82 MEV. in nickel by 7·5 MEV. deuterons, with an intensity at 90° of about 2% that of the elastic scattering. The total cross section may then be estimated in the region of 5×10^{-27} cm², which agrees to an order of magnitude with the value $2{\cdot}4 \times 10^{-27}$ cm² predicted for quadrupole excitation by equations (7) and (40).

A metastable state at 0·338 MEV. in ^{115}In has been excited by 6·9 MEV. protons (Barnes 1939) and by 16 MEV. α-particles (Risser, Lark-Horovitz and Smith 1940), the respective total cross sections being 3×10^{-29} cm² and 3×10^{-28} cm². Lawson and Cork (1940) suggest that the states involved have the spins 1/2 and 9/2 and are of opposite parity, which makes $l=5$ the only multipole operative. The cross sections predicted then by equations (7) and (40) are, for the two experiments respectively, 4×10^{-29} cm² and 3×10^{-28} cm². The exactness of this agreement must be accidental, but it is to be contrasted with the assertion of Risser, Lark-Horovitz and Smith that Weisskopf's theory predicts a cross section 10^3 times too great.

(v) *Possible Uses of Coulomb Excitation*

The inelastic scattering of charged particles has been frequently used to determine the energy of excited nuclear states. Observation has been either by direct measurement of the scattered particles, or by radioactivity of the excited state. The former is likely often to be rather difficult in the case of Coulomb excitation, because of masking by the much greater Rutherford scattering (see § 4 (iii)); however, the cross sections predicted for Coulomb excitation are not negligible.

Further information about the nature of the nuclear transition might be obtainable by measurement of other quantities, e.g. the excitation function, total cross section and angular distribution. Thus equations (7) and (27) indicate that for projectile energies well below the barrier height the partial cross section σ_l should vary as E^{2l-3} ($l>1$), down to a minimum energy $E_{\min}$ (equation (39)), below which the cross section should fall off exponentially. Therefore the excitation function might indicate the operative l value, and hence, by the selection rules (§4(ii)), the angular momentum and parity changes in the transition.

If l were known, comparison of the total cross section with the predicted (equation (7)) could yield an approximate value of the multipole moment involved $(f|Q_l|i)$.

Before such conclusions about transitions can be drawn, it would be desirable to check and perhaps improve the formulae given in this paper by detailed measurements on transitions whose characteristics are well understood.

The theory of the angular distribution (§3) is scarcely adequate yet to enable much information about the nature of the transition to be deduced from measurements; but if such detailed measurements were made in suitable cases it might be conducive to further development.

ACKNOWLEDGMENTS

We are greatly indebted to Professor H. Fröhlich for proposing this subject. We are also grateful to Professor R. E. Peierls for helpful discussions and criticisms which have resulted in a number of improvements to this paper. Professor E. Guth has kindly given permission for the use of a method of his in §2(iv).

REFERENCES

Barnes, S. W., 1939, *Phys. Rev.*, **56**, 414.
Bloch, F., 1933, *Ann. Phys., Lpz.* (5), **16**, 285.
Bush, R. R., and Fulbright, H. W., 1948, *Phys. Rev.*, **74**, 1206.
Greenlees, G. W., Kempton, A. E., and Rhoderick, E. H., 1949, *Nature, Lond.*, **164**, 663.
Guth, E., 1945, *Phys. Rev.*, **68**, 280.
Holt, J. R., and Young, C. T., 1949, *Nature, Lond.*, **164**, 1000.
Lawson, J. L., and Cork, J. M., 1940, *Phys. Rev.*, **57**, 982.
Rhoderick, E. H., 1949, *Nature, Lond.*, **163**, 848.
Risser, J. R., Lark-Horovitz, K., and Smith, R. N., 1940, *Phys. Rev.*, **57**, 355.
Sommerfeld, A., 1945, *Wellenmechanik* (New York: Ungar), Ch. 7.
Weisskopf, V., 1938, *Phys. Rev.*, **53**, 1018.

Nuclear Shell Structure and Nuclear Density

By L. M. YANG
Department of Mathematical Physics, University of Edinburgh

Communicated by M. Born; MS. received 11th December 1950

ABSTRACT. Correlations between the nuclear shell structure and the density distribution are analysed on the assumption that the former is associated with the successive appearance of particles of higher angular momentum. General requirements for the possible form of the nuclear density distribution are derived. It is shown that a reasonable density distribution conforming with the above requirements and other known facts reproduces unmistakably the ‘ magic numbers ’.

§ 1. INTRODUCTION

THE existence of a nuclear shell structure associated with particular numbers of neutrons or protons (2, 8, 20, 28, 50, 82, 126), has been derived by early workers (Elsasser 1934, Wigner 1937, Mayer 1948) from considerations of stability and abundance of nuclear systems. Further evidence was brought forward by Feenberg and Hammack (1949), Nordheim (1949), Mayer (1949), Hazel *et al.* (1949) who considered the correlations between the shell structure and the nuclear spin, magnetic moment, quadrupole moment, isomerism and β-decay. Several schemes basing on the individual particle model have been proposed to account for these numbers. These theories, though not without ambiguities in their interpretations, have, however, strengthened the concept of the nuclear shell structure.

In the present paper, it is shown that there also exist correlations between the shell structure and the nuclear density,* provided that the shell structure is associated with the appearance of particles of higher angular momentum. By virtue of this correlation, the shell structure throws light on the possible form of the nuclear density function. When a reasonable density function is assumed with adjustable constants determined by empirical facts and requirements arising from the above-mentioned association, the 'magic numbers' are unmistakably reproduced on the one hand and, on the other, it is found that for heavy nuclei ($A>50$) the thickness of the surface layer, instead of being constant and of the order of the nuclear force range, increases as $A^{1/3}$, and for light nuclei ($A<50$) the nuclear density resembles closely a gaussian function, the width of which is of course proportional to $A^{1/3}$.

§ 2. ANGULAR MOMENTUM AND NUCLEAR DENSITY

In the Thomas–Fermi model of the nucleus, the number of particles of a given angular momentum quantum number l can be calculated for an arbitrary density distribution. This idea was first successfully exploited by Fermi to calculate the electronic shell numbers in atoms. A similar investigation for the nucleus has now been suggested by Professor Born.

Let $\rho(r)$ be an arbitrary density distribution of an assembly of closely bound particles of total number n, then

$$n=\int_0^\infty \rho(r)4\pi r^2 dr.$$

The spherical symmetry of $\rho(r)$ is assumed, although this is rigorously the case only for closed shell configurations. At every point r, the associated momentum space is filled up to a maximum value $P(r)$ so that $\rho(r)=2\times(4\pi/3)\{P(r)/h\}^3$ if half spin particles are considered. It can easily be shown that the number of particles with angular momentum quantum number l among the total n particles is (Fermi 1928)

$$n_l=\frac{8}{h}(l+\tfrac{1}{2})\int_{r_1}^{r_2}[P^2(r)-(l+\tfrac{1}{2})^2(\hbar^2/r^2)]^{1/2}\,dr, \qquad \ldots\ldots(1)$$

where r_1 and r_2 must be so chosen that the integrand is real for $r_1 \leqslant r \leqslant r_2$. If the density $\rho(r)$ or $P(r)$ is known, the integral can be evaluated. In particular, the function $P(r)$ which gives $n_l=1$ will correspond to a nucleus that has just one particle in the l-state, or simpler still, the function $P(r)$ which gives $n_l = 0$, will correspond to a nucleus that has no particles in the l-state. Since the integrand in (1) is always positive, the vanishing of n_l is equivalent to

---

* After this paper was written, it was found that the problem has been tackled in a similar manner by Iwenenko and Polyjew (1950). They use only the ordinary force in calculating the nuclear density, and therefore the results are not very satisfactory.

$$\{rP(r)\}_{\max} = (l+\tfrac{1}{2})\hbar. \qquad \ldots\ldots(2)$$

If it is assumed that the successive appearance of particles of higher angular momentum is associated with the successive beginnings of new shells, then there exists a set of shell numbers for every arbitrary density distribution. In view of Fermi's success in accounting for the electronic shells and the fact that the method is independent of the force law, one can reasonably expect the validity of this method when applied to the nucleus.

§3. GENERAL REQUIREMENTS FOR THE POSSIBLE FORM OF THE NUCLEAR DENSITY DISTRIBUTION AS LIMITED BY THE SHELL STRUCTURE

We shall first state here two requirements that have to be met by any correct form of the nuclear density:

(1) The difference between the cubic roots of the successive shell numbers predicted from an arbitrary density distribution must be almost constant and equal approximately to 0·66 for large l. This is derived purely from empirical facts, viz. the last four shell numbers, as shown in the following Table[*]:

$N(l)$	28	50	82	126
$\{N(l)\}^{1/3}$	3·04	3·69	4·35	5·01
$\Delta(N^{1/3}) = \{N(l)\}^{1/3} - \{N(l-1)\}^{1/3}$		0·65	0·66	0·66

(2) The predicted proton and neutron shell numbers must be the same as empirical facts show.

It seems at first sight that this fact can hardly be reconciled with the association of the density distribution to the shell structure, since for the same closed shell, say 82, the neutron density in a nucleus of 82 neutrons is quite different from the proton density in a nucleus of 82 protons. In view of (2), the second requirement is equivalent to

$$\{rP_n(r)\}_{\max} = \{rP_p(r)\}_{\max} \qquad \ldots\ldots(3)$$

where $P_n(r)$ and $P_p(r)$ are the total local maximum momentum of the neutrons and of the protons respectively. It will be shown in the following that (*a*) the Fermi gas model of a nucleus of constant density within a certain radius and zero outside, though in conformity with the second requirement, does not quite fulfil the first, (*b*) the modifications introduced by considering the Coulomb interaction make the form of the density distribution even worse, in so far as the first requirement is concerned, and (*c*) the thickness of the surface layer of all nuclei is proportional to the nuclear radius instead of being constant.

Let the total density be $\rho(r)$, and the partial densities of the protons and the neutrons be $\rho_p(r)$ and $\rho_n(r)$ respectively, then for the Fermi gas model one has

$$\left.\begin{aligned} \rho(r) = \rho_n(r) + \rho_p(r) &= (4\pi r_0^3/3)^{-1} \quad \text{for } r \leqslant R \\ &= 0 \qquad\qquad\quad \text{for } r > R \end{aligned}\right\}. \qquad \ldots\ldots(4)$$

---

[*] For the reason why the first few numbers do not show this regularity, see § 5 (*d*).

The weakness of Coulomb interaction permits us to assume

$$\rho_{\mathrm{n}}(r)/\rho_{\mathrm{p}}(r)=N/Z,\ A=N+Z \qquad \ldots\ldots(5)$$

where N and Z are the total numbers of neutrons and protons in a nucleus of mass number A, r_0 is the radius of a sphere occupied on the average by each nucleon, the exact value of which is not essential for the present purpose as r_0 will only appear as a proportional constant, and $R=A^{1/3}r_0$ is the nuclear radius. From (2) it follows

$$\{rP_{\mathrm{n}}(r)\}_{\max}=RP_{\mathrm{n}}(R)=(l+\tfrac{1}{2})\hbar. \qquad \ldots\ldots(6)$$

Since

$$P_{\mathrm{n}}(R)=\left(\frac{9\pi}{4}\frac{N}{A}\right)^{1/3}\frac{\hbar}{r_0},$$

(6) becomes $N^{1/3}=(2l+1)/(18\pi)^{1/3}$. Thus $\Delta(N^{1/3})=(4/9\pi)^{1/3}=0{\cdot}52$. The constancy of $\Delta(N^{1/3})$ is obtained here, but its value 0·52 is too small compared with the empirical 0·66. The second requirement is satisfied by (4), as the neutron shell numbers predicted are independent of A; the dependence of R on A just cancels that of $P_{\mathrm{n}}(R)$ on A. Statement (*a*) is therefore proved. Statement (*b*) is obvious since one notices in (6) that increasing density towards the rim due to Coulomb effect as estimated by Feenberg (1941) tends to increase $\{rP_{\mathrm{n}}(r)\}_{\max}$, i.e. $\{rP_{\mathrm{n}}(r)\}_{\max}=(CN)^{1/3}$ where $C>9\pi/4$, and therefore $\Delta(N^{1/3})$ becomes still smaller. Statement (*c*) will be considered in the next section.

It may be added that the two requirements stated above apply essentially to nuclear density of heavy nuclei, since both large angular momentum states and large neutron excess occur only in these nuclei.

§4. SHELL NUMBERS REPRODUCED FROM NUCLEAR DENSITY

In view of the fact that the binding energy per nucleon is almost constant and the nuclear radius is proportional to $A^{1/3}$, we assume a density function of the form

$$\left.\begin{aligned}\rho(r)&=\rho_0 && \text{for } r\leqslant R_0\\ &=\rho_0\exp\left[-\{(r-R_0)/a\}^2\right] && \text{for } r>R_0\end{aligned}\right\} \qquad \ldots\ldots(7)$$

where a, ρ_0 and R_0 are functions of A to be determined by the two requirements in §3. The radius R is defined by $R=R_0+a$, and the empirical relation $R=A^{1/3}r_0$ requires that

$$R_0+a=r_0A^{1/3}. \qquad \ldots\ldots(8)$$

To a first approximation, we also assume $\rho_{\mathrm{n}}(r)=(N/A)\rho(r)$, $\rho_{\mathrm{p}}(r)=(Z/A)\rho(r)$. The normalization of $\rho(r)$ gives

$$A=\int_0^\infty \rho(r)4\pi r^2\,dr=\frac{4\pi}{3}\rho_0a^3\left[\frac{3\sqrt{\pi}}{4}\left\{1+2\left(\frac{R_0}{a}\right)^2\right\}+\left(\frac{R_0}{a}\right)\left\{3+\left(\frac{R_0}{a}\right)^2\right\}\right]. \qquad \ldots\ldots(9)$$

Now consider the neutron shell first; we have

$$\{rP_{\mathrm{n}}(r)\}^3{}_{\max}=3\pi^2(r_{\mathrm{m}}\hbar)^3\rho_0\frac{N}{A}\exp\left\{-\left(\frac{r_{\mathrm{m}}-r_0}{a}\right)^2\right\}=(l+\tfrac{1}{2})^3\hbar^3 \qquad \ldots\ldots(10)$$

where r_{m} is the value of r at which $\{r\rho_{\mathrm{n}}(r)\}$ is a maximum (see Figure).

$$r_{\mathrm{m}}=\{R_0+(R_0{}^2+6a^2\}^{1/2}/2$$

or

$$\frac{R_0}{a}=\frac{1}{\sigma}-\frac{3}{2}\sigma,\quad \sigma=\frac{a}{r_{\mathrm{m}}}. \qquad \ldots\ldots(11)$$

From (10) and (11), it follows that

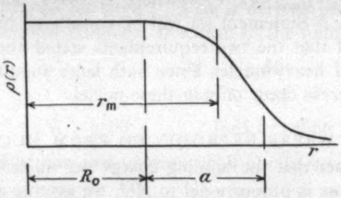
$$N=\frac{(2l+1)^3}{18\pi}f(\sigma)\exp\left(\tfrac{3}{2}\sigma\right)^2 \qquad \ldots\ldots(12)$$

$$\text{where } f(\sigma)=\left\{1+\frac{3\sqrt{\pi}}{2}\sigma\left(1-\tfrac{5}{2}\sigma^2+\tfrac{9}{4}\sigma^4\right)-\tfrac{3}{2}\sigma^2\left(1-\tfrac{3}{2}\sigma^2+\tfrac{9}{4}\sigma^4\right)\right\}.$$

It is evident that for heavy nuclei, our two requirements can be met by assigning a constant value to σ. The relation $\Delta(N^{1/3})=0{\cdot}66$ requires that $\exp(3\sigma/2)^2 f(\sigma)=2{\cdot}1$ which gives $\sigma=0{\cdot}38$. With the parameter σ so determined, the neutron shell numbers calculated from (12) agree remarkably well with the last four magic numbers. The results are as follows:

l	=	3	4	5	6
$N(l)$	=	27·1	49·5	81·5	125·2
Next integer	=	28	50	82	126

The proton shell numbers come out the same provided that the same value of σ is taken. With $\sigma=0{\cdot}38$, we have from (11) $R_0/a=2{\cdot}06$. Thus for all nuclei with A greater than 50, the thickness of the surface layer increases proportionally with the linear dimension of the main body of constant density. Statement (*c*) in §4 is therefore proved. The other constants are uniquely determined from (8) and (9):



$$R_0=0{\cdot}673r_0A^{1/3},\ a=0{\cdot}327r_0A^{1/3},\ \rho_0=1{\cdot}04/(4\pi r_0^3/3). \qquad \ldots\ldots(13)$$

The density at the centre of the nuclei ρ_0 differs little from the usual value $(4\pi r_0^3/3)^{-1}$.

As the number of particles decreases, the central part of constant density shrinks gradually, so that for A less than 50, the nuclear density can reasonably be expected to resemble the form of a gaussian function. With $R=0$ in (7), we have from (11) and (8)

$$R_0=0,\ a=r_0'A^{1/3},\ \sigma^2=2/3\ (\sigma=0{\cdot}816),\ \rho_0=0{\cdot}752/(4\pi r_0'^3/3) \qquad \ldots\ldots(14)$$

where r_0' may be slightly larger than r_0, as the nucleons are less firmly bound in light nuclei. In fact, as r_0 and r_0' appear only as proportional constants their values can be assigned separately for each full shell in order to obtain best fit with the true densities. The neutron (or proton) shell numbers as calculated from (12) are

l	=	0	1	2
$N(l)$	=	1·55	7·16	19·66
Next integer	=	2	8	20

The empirical shell numbers are again obtained. It should be noted that the value of σ is uniquely determined for $R=0$. Hence the assumption of a gaussian density function alone suffices to predict correctly the first three shell numbers. This is really what one ought to expect, as the early theories based on the individual particle energy levels agree with each other up to the shell number 20 (Feenberg *et al.* 1949). This also indicates that the oscillator potential is perhaps a good approximation for light nuclei.

§ 5. DISCUSSION

The fact that the shell numbers can be reproduced so accurately by taking a reasonable form of the density distribution, limited further by the two requirements stated in § 3, leads one to expect that the assumption on the connection between shell structure and angular momentum has meaning. By exploiting this correlation more can be inferred concerning both of them. We wish to mention further the following:

(*a*) In § 4, different values of σ have to be assigned for heavy and light nuclei. This situation is presumably created by the Coulomb repulsion, in which case the value of σ should vary gradually from 0·816 for the inner shells to 0·38 for the outer shells, and the values of R_0 and ρ_0 should also increase gradually from those given in (14) to those given in (13). A transition must exist between the two sets of constants, and this seems to take place between the shell numbers 20 and 28. Detailed calculation of the nuclear densities is required to confirm this assertion. In support of this point of view, the fact may be cited that '20' is the largest shell for which a full proton shell can co-exist with the same full neutron shell—an indication that the Coulomb effect starts to be appreciable from the shell '20' on.

(*b*) The existence of a central part of almost constant density in heavy nuclei is in part at least due to the effect of the Coulomb repulsion, without which the density distribution would be almost gaussian for all existing nuclei.

(*c*) One of the consequences arising from our result of the proportionality of the thickness of the surface layer to $A^{1/3}$ is that the surface energy estimated in the usual manner no longer varies as $A^{2/3}$ but as A. This does not, however, constitute a serious objection to the theory suggested, for terms in the surface energy proportional to $A^{1/3}$ also appear in the improved Thomas–Fermi model (von Weizsäcker 1937) where the kinetic energy density at a given point depends not only on ρ but also on grad ρ. Besides, terms proportional to $A^{2/3}$ may arise from the discreteness of the particle energy levels (Rosenfeld 1948). It is true that the presence of a surface region and therefore of a deficiency of the binding energy (the surface energy) is definitely required both on theoretical grounds and by empirical evidence; the semi-empirical result that the surface energy should be proportional to $A^{2/3}$ alone, a simple relation taken over from the purely classical picture, cannot be regarded as final but only represents one of the possibilities of a simple assumption to fit experimental facts (mass defect and nuclear fission).

(*d*) According to equations (12) and (14), we should have for the light nuclei $\Delta(N^{2/3})=0{\cdot}77$. This regularity which has been observed for heavy nuclei, does not appear from the empirical shell numbers of light nuclei (2, 8, 20); the reason is that the way in which (1) is derived is semi-classical. The conversion from a (classical) continuous angular momentum spectrum to a (quantal) discrete one is not without arbitrariness, which, though negligible for high quantum number l, is appreciable for small l. The rule $\Delta(N^{1/3}) = \text{const.}$ derived from (1) is therefore only a classical approximation that cannot be expected to be rigidly followed down to empirical shell numbers corresponding to small l, though the rule can be satisfied by properly choosing numbers between each shell number and its next smaller integer.

ACKNOWLEDGMENTS

The author is greatly indebted to Professor Max Born for his advice and encouragement and, in particular, for his inspiring suggestion of considering the Thomas–Fermi model of nuclei. He is also indebted to the University of Edinburgh for the tenure of a research fellowship.

REFERENCES

Elsasser, W., 1934, *J. Phys. Radium*, **5**, 625.
Feenberg, E., 1941, *Phys. Rev.*, **59**, 593.
Feenberg, E., and Hammack, K. C., 1949, *Phys. Rev.*, **75**, 1877.
Feenberg, E., Hammack, K. C., and Nordheim, L. W., 1949, *Phys. Rev.*, **75**, 1968.
Fermi, E., 1928, *Z. Phys.*, **48**, 73.
Haxel, O., Jensen, J. H. D., and Suess, H. E., 1949, *Phys. Rev.*, **75**, 1766.
Iwanenko, D., and Polyjew, W., 1950, *Dokl. Akad. Nauk, U.S.S.R.*, **70**, 605.
Mayer, M. G., 1949, *Phys. Rev.*, **75**, 1766; 1948, *Ibid.*, **74**, 235.
Nordheim, L. W., 1949, *Phys. Rev.*, **75**, 1894.
Rosenfeld, L., 1948, *Nuclear Forces* (Amsterdam: North Holland Publishing Co.).
von Weizsäcker, C. F., 1937, *Die Atomkerne* (Leipzig: Akademische Verlagsgesellschaft).
Wigner, E., 1937, *Phys. Rev.*, **51**, 947.

A Method of Calibrating Neutron Sources Absolutely

By D. J. LITTLER
Atomic Energy Research Establishment, Harwell, Berks.

MS. received 31*st October* 1949, *and in final form* 27*th March* 1951

ABSTRACT. A method has been developed for calibrating neutron sources absolutely, using the Graphite Low Energy Experimental Pile. The method used is to convert the measurement of the number of neutrons emitted by a source to a determination of the absolute beta activity of an irradiated sodium or phosphorus sample.

One source calibrated by this method, which contained 1,290 mg. of radium in the form of sulphate, mixed with 12 gm. of beryllium, was found to have a neutron strength of $9{\cdot}3 \times 10^6$ n/second ($\pm 4\frac{1}{2}\%$).

§ 1. INTRODUCTION

The method of calibrating neutron sources described here avoids the difficulties usually encountered in counting either fast or slow neutrons. Since only one absolute measurement need be made, use is made of a pile in this experiment, to convert the measurement of the number of neutrons coming from a neutron source to a determination of the number of β-particles emitted by a radioactive source. If this is done, the coincidence or defined solid angle techniques for the absolute measurement of β-activity may be used and these methods are capable of yielding high accuracy of the order of within $\pm 2\%$.

The measurement of absolute β-activity will not be described in this paper, and the reader is referred to a paper of Putman (1950) which gives a detailed account of the subject.

§ 2. THEORY OF THE METHOD

If a neutron source is introduced into a steadily running pile, it will have two effects on the pile: the material from which it is made will absorb thermal neutrons from the pile, and the source itself will give out fast neutrons to the pile. The pile power will rise if the source gives out more neutrons than it absorbs, and the power will fall if the opposite state of affairs prevails.

It is customary, when discussing changes in the pile brought about by introducing material into it, to consider the change in the effective reproduction constant of the pile. The effective reproduction constant of the pile is defined as the number of thermal neutrons born and remaining in the pile for each thermal neutron absorbed in the pile. Evidently, for a steadily running pile, the effective reproduction constant k_e must be unity; if k_e is less than unity the pile will converge, and if it is greater than unity, the pile will diverge.

By the use of a diffusion theory, the effect on k_e of introducing a neutron source into the pile has been worked out. This theory is given in an Appendix, and shows that the change δ in k_e is given by

$$\delta = A\{\omega S/\rho_S - B\} \qquad \ldots\ldots(1)$$

where A is a constant, provided that the source is always put in the same place in the pile, B is another constant proportional to the neutron absorption cross section of the source, and S is the strength of the source (number of neutrons emitted by it per second); ρ_S is the value of the thermal neutron density at the source position when the source is absent from the pile, and ω is a combination of pile constants.

The numerical value of ω is 0·875, when the neutron source under consideration is a radium beryllium (α, n) source, but the value of ω is not very critically dependent on the energy of the neutrons emitted by the source. In fact, the only pile constant which is really important in the evaluation of ω is the value of the resonance escape probability. This is the probability that a fast neutron will escape resonance capture in ^{238}U, and become a thermal neutron. The quantity ω is a kind of weighting factor which allows for the fact that fast neutrons have to escape the resonance trap before they can give rise to thermal fission, whereas thermal neutrons absorbed by the source could alternatively have caused thermal fission.

If the power level of the pile is measured with a boron trifluoride ionization chamber which is kept in a fixed position in the pile, the ion current from this chamber will always be proportional to ρ_S. As explained in the next section, the ion current is found by measuring the voltage V developed across a stable resistor in series with it; then $\rho_S = CV$, where C is a constant. Thus (1) may be written $\delta = A\{\omega S/CV - B\}$.

If a series of measurements of δ be made for a number of values of V, and δ plotted against $1/V$, the slope m of the straight line so obtained will be given by

$$m = A\omega S/C. \qquad \ldots\ldots(2)$$

Now suppose a measurement of δ is made with some substance on which an absolute determination of the β-activity produced by neutron absorption can also be made. (The element must be a single isotope element, such that its activation cross section is the same as its absorption cross section. Sodium and phosphorus were two such elements used.) If the value of δ for this calibrating substance be δ^*, then δ^* is given (see Appendix) by

$$\delta^* = -A\alpha \qquad \text{......(3)}$$

where α is a quantity which is proportional to the absorption cross section of the calibrating element, and in fact the rate of neutron absorption by the element is $\rho_S\alpha$; it is also assumed that the measurement of δ is made with the element in the same position in the pile as was occupied by the neutron source.

Suppose also that by absolute β counting we can determine N, the rate of neutron absorption by the calibrating substance when the value of ρ_S is ρ_0, and the value of V is V_0. Then

$$N = \alpha\rho_0 = \alpha C V_0. \qquad \text{......(4)}$$

Combining (2), (3) and (4) we have,

$$S = Nm/\omega\,|\delta^*|\,V_0. \qquad \text{......(5)}$$

Thus the source strength is determined in terms of N, which is itself derived from absolute measurements of β-activity.

It should be noted that it is not necessary to make absolute measurements of δ; it is sufficient to measure any quantity which is proportional to δ.

§3. MEASUREMENT OF THE CHANGE IN k_e WITH A RADIUM BERYLLIUM SOURCE

The source calibrated in this experiment was housed in a graphite container which could be driven from the outside of the pile shield to the centre of the pile by means of an electro-mechanical system. The source was moved between these two positions in a regular manner so as to produce a periodic modulation of the pile power (the source remained in each position for 20 seconds and took 2 seconds to move from one position to the other).

Figure 1 is a block diagram of the electrical apparatus used in the measurements. The main control unit governs the motion of the source in the pile. The current from the boron trifluoride ionization chamber consists of two parts: (*a*) A steady current, which produces a voltage across the resistor in series with the chamber. This voltage is balanced by the 'backing-off' voltage, and the backing-off voltage is a measure of the chamber current. (*b*) A periodically varying current due to the periodic modulation of the pile by the neutron source. The voltage that this produces across the resistor is amplified by the D.C. amplifier.

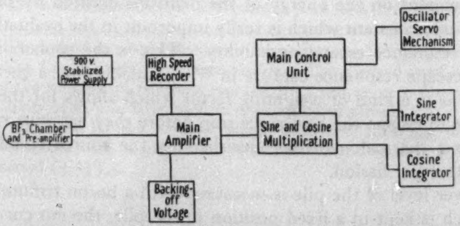

Oscillator Servo Mechanism
900 v. Stabilized Power Supply
High Speed Recorder
Main Control Unit
Sine Integrator
BF3 Chamber and Pre-amplifier
Main Amplifier
Sine and Cosine Multiplication
Cosine Integrator
Backing-off Voltage

Figure 1. Schematic diagram of electrical equipment.

The voltage output from the amplifier is multiplied separately by sine and cosine factors of the same periodicity as the motion of the sample (this is done by having sliders moving with simple harmonic motion across resistor cards connected to the output of the amplifier); it is then fed into two velodyne integrators and integrated for one complete period.

The integrated answers are thus proportional to the two fundamental components in the Fourier analysis of the pile modulation. If we consider only the first harmonic components in the pile modulation, it may be shown that the percentage modulation produced by the neutron source is proportional to the change in k it would produce if placed at the centre of the pile (Weinberg and Schweinler 1948). Thus the integrator answers are a measure of δ.

Two runs were made with the neutron source, and twenty observations were taken at every power level used. The mean values obtained at each power level for both runs are plotted as a function of $1/V$ in Figure 2. In order to indicate the sensitivity of the method, the absolute values of the change in k recorded are shown on the vertical scale. The abscissae are plotted in terms of the inverse of the 'backing-off' voltage.

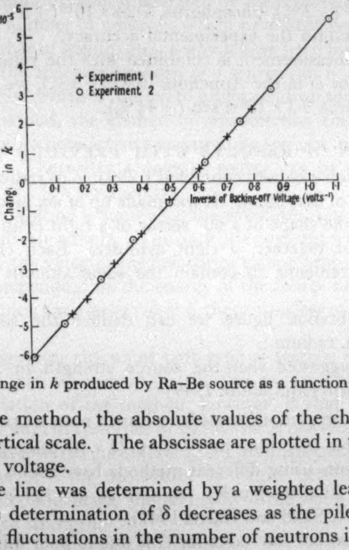


Figure 2. Change in k produced by Ra–Be source as a function of pile power.

The slope of the line was determined by a weighted least squares method (the accuracy of the determination of δ decreases as the pile power is reduced, because of statistical fluctuations in the number of neutrons in the pile), and was found to be $m = 10{\cdot}84 \times 10^{-5}$ volts ($\pm 1\frac{1}{2}\%$).

There was a possibility that some of the gamma-rays produced in the pile might eject photo-neutrons from the source, thus giving an erroneous value for its strength. The gamma flux in the pile was therefore measured, and the (γ, n) effect was found to be negligible.

§4. MEASUREMENTS WITH SODIUM CARBONATE AND PHOSPHORUS

In much the same way as described in the previous section, measurements of δ were made with samples of both sodium carbonate and phosphorus. Since we are only interested in determining those neutrons which are absorbed to produce ^{24}Na and ^{32}P, corrections have to be made for the following effects: (*a*) neutrons absorbed by carbon and oxygen in the sodium carbonate, (*b*) neutrons absorbed by chemical impurities in both samples, (*c*) fast neutrons which produce (n, p) and (n, α) reactions in both samples. These corrections were not large for the sodium carbonate, but allowance had to be made for a number of impurities in the red phosphorus.

The sodium carbonate and phosphorus samples were also irradiated at the centre of the pile and subsequently measured to obtain the absolute amount of ^{24}Na and ^{32}P activity induced (Putman 1950).

These two samples were used because the absolute counting is based on two entirely different methods; for ^{24}Na a coincidence technique is used and for ^{32}P defined solid angle counting is used.

A large number of measurements of β-activity was made on sodium carbonate, but only one measurement of β-activity was made on the phosphorus sample. For this reason, the phosphorus measurement was not used in finally computing the strength of the neutron source. The values of $N/|\delta^*|V_0$ obtained were: sodium, $7{\cdot}51\times10^{10}$ ($\pm3\%$), phosphorus, $7{\cdot}24\times10^{10}$ ($\pm5\%$); it can be seen that the results agree within the experimental accuracy.

If the sodium measurement is combined with the value obtained for *m*, and the value quoted for ω in the Appendix ($0{\cdot}875\pm3\%$), we have for the strength of the neutron source $9{\cdot}3\times10^6$ n/sec. ($\pm4\frac{1}{2}\%$).

§ 5. COMPARISON OF RESULTS WITH PREVIOUS DETERMINATIONS

The source which was used contained 1,290 mg. of radium as radium sulphate mixed with 12 gm. of beryllium. It was made up of six pieces of the same shape; each piece was in the shape of a 60° sector of a right cylinder, the six 'cheeses' forming, when put together, a right cylinder. Each cheese was found, by gamma-ray measurements, to contain the same amount of radium to better than 1%.

From the calibration figure we can deduce the source strength to be $0{\cdot}72\times10^7$ n/sec/gm. radium.

It has been suggested that the source strength in n/sec/gm. Ra can be expressed in the form (McCallum 1949)

$$C\,(\text{mass beryllium})/(\text{mass beryllium}+\text{mass of radium salt}).$$

Thus, for the source described here, $C=0{\cdot}83\times10^7$ n/sec/gm. Ra.

Other experiments using different methods have found values for *C* ranging from $1{\cdot}1$ to $1{\cdot}6\times10^7$ n/sec/gm. Ra (Agnew *et al.* 1947, Bauer *et al.* 1943, Bretscher *et al.* 1944, Seidl and Harris 1947).

It is believed that our low value for *C* is due to poor mixing of the beryllium and radium sulphate in some of the cheeses.

The relative neutron strengths of the cheeses were obtained by placing each cheese in turn at the centre of a large graphite block, and observing the neutron flux at another point in the block with a BF$_3$ proportional counter.

The results are given in the Table in which the last two lines show the results of such intercalibrations performed both before and after the absolute calibration.

Relative Strengths of Cheeses

Cheese no.	1	2	3	4	5	6
Early measurement	175	137	151	99	135	108
Before calibration	171	136	153	99	134	112
After calibration	173	135	158	96	133	110

The second line gives the relative strengths obtained several years ago by another experimenter. Since all the measurements are only accurate to within $\pm 2\%$, these results show that the neutron strengths of the cheeses do not vary appreciably with time.

It will be seen that there is a wide range of values for the neutron strengths of the cheeses. If we take the results from cheese No. 1 only, we have $C = 1{\cdot}1 \times 10^7$ n/sec/gm. Ra. This figure now comes within the range of values found by other experimenters.

§ 6. DISCUSSION OF PRESENT METHOD

The method of calibrating neutron sources described here is as accurate as most of the other methods which have been published. It is not so accurate as the method proposed by Glueckauf and Paneth (1938), in which the amount of helium liberated from beryllium in a photo-neutron source is determined. In this latter method the amount of helium liberated by the reaction $^9\text{Be} + h\nu \rightarrow 2\,^4\text{He} + \text{n}$ is measured, and the number of neutrons which have been emitted by the source is one half of the number of helium atoms which have been formed. It is claimed that by this method the absolute strength of a source can be determined to 1%. It is, however, a method which is limited to photo-neutron sources.

In the present method, the change in reproduction constant produced by sodium, and the absolute beta-activity acquired by sodium in the pile, need only be determined once. Thereafter a neutron source can be calibrated to better than 5% in a matter of a few hours only.

The theory of the method is somewhat involved (see Appendix), but the final result obtained is relatively simple. The only pile constant which need be known with high accuracy is the value of the resonance escape probability. The theory is only very slightly dependent on the energy of the source neutrons, and since a nuclear pile is used which has a large volume, there is no loss of neutrons by leakage to be considered.

It has an advantage over the water tank type of method (Agnew *et al.* 1947, Bauer *et al.* 1943) in that no neutron distribution measurements need be made.

One serious limitation of the method, however, is that the neutron source being calibrated must emit of the order of 10^7 neutrons per second, or the accuracy of the determination is lowered considerably. This arises because the accuracy of each individual value of δ for the source is mainly determined by statistical fluctuations in the pile, and hence the accuracy of the slope of the line $(\delta, 1/V)$ depends on the source strength.

§ 7. CONCLUSION

The method of calibrating neutron sources described here is capable of yielding accuracy of the order of $\pm 5\%$. It is, of course, only of value to anyone who has access to a nuclear pile. But if a neutron source which is reproducible, such as that described by Bretscher *et al.* (1949) or by Curtiss and Carson (1949) be calibrated by this method, then this source could serve as a standard neutron source.

ACKNOWLEDGMENTS

The author would like to express his thanks to Miss A. M. Wildblood and Mr. J. L. Putman of Atomic Energy Research Establishment, who did the absolute counting, to the staff of the Analytical Services section of C.C.I., Chatham, who did the chemical analyses of the sodium carbonate and phosphorus samples, and to Messrs. W. G. L. Brownrigg and E. E. Lockett who assisted in taking the pile measurements. The author is also indebted to the Director of the Atomic Energy Research Establishment for permission to publish this paper.

REFERENCES

AGNEW, H. M., *et al.*, 1947, *Manhattan District Declassification Committee Report* 878.
BAUER, S., *et al.*, 1943, *British Report* 134.
BRETSCHER, E., *et al.*, 1944, *British Report* 382.
BRETSCHER, E., *et al.*, 1949, *Proc. Roy. Soc.* A, **196,** 436.
CURTISS, L. F., and CARSON, A., 1949, *Phys. Rev.*, **76,** 1412.
GLUECKAUF, E., and PANETH, F., 1938, *Proc. Roy. Soc.* A, **165,** 229.
PUTMAN, J. L., 1950, *Brit. J. Radiol.*, **23,** 46.
MCCALLUM, K. J., 1949, *Nucleonics*, **5,** 11.
SEIDL, F. G. P., and HARRIS, S. P., 1947, *Rev. Sci. Instrum.*, **18,** 897.
WEINBERG, A. M., and SCHWEINLER, H. C., 1948, *Phys. Rev.*, **74,** 851.

APPENDIX

THEORY OF THE EXPERIMENT USING A THREE-GROUP MODEL

The three neutron groups considered are: (*a*) Neutrons with energies between those emitted from the source and those emitted in fission. Let the density of these neutrons, a function of position in the pile, be denoted by s. (*b*) Neutrons with energies between fission energy and the energy at which capture takes place in the ^{238}U resonance—density f. (*c*) Neutron with energies below the ^{238}U resonance—density ρ. When the neutron source is not in the pile, we are only concerned with the last two groups, and so we will consider this case first in order to derive a relationship between f and ρ.

If the pile is running steadily, we have for a unit volume per unit time for both neutron groups: Production = Capture + Outflow.

For neutrons in both groups, the term representing outflow will be the divergence of a current. It we suppose the neutrons to obey a diffusion theory, then we can write the current as a diffusion coefficient, D, times the gradient of a neutron density, and thus the outflow term is of the form

$$\text{Outflow} = -D\nabla^2 \text{(density)}.$$

Suppose that k_1 is the reproduction constant of the pile in this case (the number of thermal neutrons obtained for each thermal neutron absorbed in the pile), and that τ_1 and τ_2 are the mean lifetimes of neutrons in groups (*c*) and (*b*) respectively. Then using the continuity equation, we have,

$$D_1\nabla^2\rho - \rho/\tau_1 = -pf/\tau_2 \qquad \ldots\ldots(1)$$

and

$$D_2\nabla^2 f - f/\tau_2 = -k_1\rho/p\tau_1 \qquad \ldots\ldots(2)$$

where D_1 and D_2 are the diffusion coefficients for the two groups, and p is the probability that a fast neutron will escape resonance capture. In equation (1) the term on the right-hand side represents the rate of production of thermal neutrons, since for each fast neutron dying only p thermal neutrons are produced. Similarly in equation (2) k_1/p fast neutrons are produced by the death of each thermal neutron.

Equations (1) and (2) may also be written as

$$\nabla^2\rho - \rho/L^2 = -pf\tau_1/L^2\tau_2 \qquad \ldots\ldots(3)$$

and

$$\nabla^2 f - f/L_s^2 = -k_1\rho\tau_2/pL_s^2\tau_1 \qquad \ldots\ldots(4)$$

where $D_1\tau_1=L^2$ and $D_2\tau_2=L_s^2$. L is called the diffusion length, and L_s the slowing-down length, for neutrons in the pile.

By eliminating either f or ρ from the two second-order equations, we find that both f and ρ satisfy the same fourth order equation:

$$\left[\left(\nabla^2-\frac{1}{L^2}\right)\left(\nabla^2-\frac{1}{L_s^2}\right)-\frac{k_1}{L^2L_s^2}\right]Z=0. \qquad \ldots\ldots(5)$$

The operator in brackets may be factorized to give

$$(\nabla^2+K^2)(\nabla^2-\eta^2)Z=0. \qquad \ldots\ldots(6)$$

Thus f and ρ are linear combinations of the solutions of

$$\nabla^2Z=-K^2Z, \qquad \nabla^2Z=\eta^2Z.$$

If we now solve the latter two equations for a cylindrical pile with axial symmetry (coordinates z, r), and then insert the boundary conditions that the density of neutrons must be finite throughout the pile, and must fall to zero at $z=\frac{1}{2}h$ (h being the effective length of the pile) and $r=R$ (R being the effective radius of the pile), we find that ρ and f are given by

$$\rho=A_1\cos(\pi z/h)\,.\,J_0(j_{01}r/R) \qquad \ldots\ldots(7)$$

$$f=A_2\cos(\pi z/h)\,.\,J_0(j_{01}r/R) \qquad \ldots\ldots(8)$$

where $\pi^2/h^2+j_{01}^2/R^2=K^2$, and A_1 and A_2 are arbitrary constants. By substituting for ρ and f in equation (4), we also have $A_2=k_1\tau_2A_1/p(K^2L_s^2+1)\tau_1$.

Thus before the introduction of the neutron source, ρ and f are everywhere related by the equation

$$f=k_1\tau_2\rho/p(K^2L_s^2+1)\tau_1. \qquad \ldots\ldots(9)$$

K^2 is a quantity which determines the size of the pile and is called the Laplacian. In the case of Gleep, $K^2=0{\cdot}92\pm0{\cdot}04\,\mathrm{m}^{-2}$.

We now consider the case in which the neutron source has been introduced into the pile. We assume that there is a small cavity at the centre of the pile ($z=0$, $r=0$), and that before the neutron source was introduced, the cavity was empty. (With the cavity empty, equations (7), (8) and (9) describe the state of affairs everywhere in the pile except in the cavity itself.)

We suppose that when the neutron source was introduced into the cavity, the reproduction constant was changed to k_2 so that the pile was again running steadily. The neutron densities are now assumed to be ρ_1, f_1 and s, and the equations governing these densities are,

$$D_1\nabla^2\rho_1-\rho_1/\tau_1=-pf_1/\tau_2 \qquad \ldots\ldots(10)$$

$$D_2\nabla^2f_1-f_1/\tau_2=-k_2\rho_1/p\tau_1-s/\tau_3 \qquad \ldots\ldots(11)$$

$$D_3\nabla^2s-s/\tau_3=0 \qquad \ldots\ldots(12)$$

where D_3 and τ_3 are the diffusion coefficient and mean lifetime of neutrons in group (a).

If we now multiply equation (1) by ρ_1, (10) by ρ, subtract the two equations, and then integrate over the whole volume of the pile, we have

$$-D_1\int(\rho_1\nabla^2\rho-\rho\nabla^2\rho_1)dV=(p/\tau_2)\int(f\rho_1-f_1\rho)dV.$$

We may transform the left-hand side into a surface integral by the use of Green's theorem, and remembering that ρ and ρ_1 are both zero at the outer boundary of the pile, we are left with only the surface integral over the cavity:

$$(p/\tau_2)\int(f\rho_1-f_1\rho)dV=-D_1\int(\rho_1\,\mathrm{grad}\,\rho-\rho\,\mathrm{grad}\,\rho_1)dS.$$

Now $-\int D_1\,\mathrm{grad}\,\rho\,dS$ represents the net inflow of neutrons into the cavity per unit time, so that if the cavity is sufficiently small for ρ to be regarded as constant over its surface and equal to ρ_c,

$$(p/\tau_2)\int(f_1\rho-f\rho_1)dV=\rho_c\times\text{rate of thermal neutron absorption in cavity when source is present,}$$

since the net inflow into the cavity is zero when it is empty. Thus, if T is the rate of absorption of thermal neutrons by the source, then

$$(p/\tau_2)\int(f_1\rho-f\rho_1)dV=\rho_c T. \qquad \ldots\ldots(13)$$

By a similar treatment, and using equations (2) and (11), we have

$$(1/p\tau_1)\int(k_2f\rho_1-k_1f_1\rho)dV+\int sf/\tau_3dV=f_cF \qquad \ldots\ldots(14)$$

where f_c is the value of f at the cavity, and F is the rate of absorption of neutrons in group (*b*) by the source.

Eliminating $\int f_1\rho dV$ from (13) and (14), and also using (9), we have

$$(k_2-k_1)\int\rho\rho_1dV+p\tau_1\int s\rho/\tau_3dV=\rho_c\tau_1\{(K^2L_s^2+1)T+pF\}. \qquad \ldots\ldots(15)$$

Now s is determined by equation (12), and since s/τ_3, the rate of dying of these neutrons at any point, must be proportional to S the strength of the neutron source, we may write

$$s/\tau_3=S\phi(z,r) \qquad \ldots\ldots(16)$$

where $\phi(z,r)$ is a function of the cylindrical coordinates in the pile, and is determined from equation (12). Furthermore we may write equation (7) as $\rho=\rho_c\cos(\pi z/h)\,.\,J_0(j_{01}r/R)$ since the cavity is at the centre of the pile.

Thus the term involving S in equation (15) may be written as

$$p\tau_1S\rho_c\int\phi(z,r)\cos(\pi z/h)\,.\,J_0(j_{01}r/R)dV.$$

This integral has been evaluated numerically, and we will write it in the form $(1-Y)$. Assuming that the neutrons emitted from a radium beryllium source are of energy 5 MEV. and those emitted in fission of energy 2 MEV., Y is evaluated as 0·002.

It may be shown that if only a two-group model is used (neutrons in groups (*a*) and (*b*) considered together), then $Y=0$. Thus the small value of Y obtained when a three-group model is used shows that the experimental results do not depend very critically on the energy of the source neutrons.

In equation (15) we may also write $\int\rho\rho_1dV=\int\rho^2dV$, since $\rho=\rho_1$ over almost the whole of the pile if the introduction of the source may be considered as only a small perturbation.

Thus from equation (15) we now have

$$(k_2-k_1)=\frac{(K^2L_s^2+1)\tau_1\rho_c}{\int\rho^2\,dV}\left\{T+\frac{p}{(1+K^2L_s^2)}[F-S(1-Y)]\right\}.$$

We may also write $T=\alpha\rho_c$, $F=\beta\rho_c$ so that

$$(k_2-k_1)=\frac{(K^2L_s^2+1)\tau_1\rho_c^2}{\int\rho^2\,dV}\left\{\alpha+\frac{p}{(1+K^2L_s^2)}\left[\beta-\frac{S(1-Y)}{\rho}\right]\right\}.$$

Now for a fixed position of the cavity in the pile $\rho_c^2/\int\rho^2 dV$ will be constant, independent of the level at which the pile is running, and so we may write $(k_2-k_1)=A\{\alpha+W[\beta-S(1-Y)/\rho_c]\}$ where $W=p/(1+K^2L_s^2)$ and A is a constant.

To simplify the expression slightly we also write $\alpha+W\beta=B$, another constant. Then $(k_2-k_1)=A\{B-WS(1-Y)/\rho_c\}$.

This equation tells us the change in the lattice reproduction constant which would have to be made in order that the pile should continue running steadily after introducing the neutron source. In practice this change cannot be made, and introduction of the neutron source will cause a change in the effective reproduction constant of the pile. (This effective reproduction constant is equal to the value of the lattice reproduction constant minus a term giving the leakage of neutrons from the pile; the effective reproduction constant must be unity for a steadily running pile.)

The change δ in the effective reproduction constant, on introducing the source into the pile, will be given by

$$\delta=A\{W(1-Y)S/\rho_c-B\}. \qquad \ldots\ldots(17)$$

If we consider the change δ^* produced by introducing a sodium sample into the cavity, and write T_1 the number of thermal neutrons absorbed per second by sodium as $\alpha_1\rho_c$, and F_1 the number of fast neutrons absorbed per second by sodium as $\beta_1\rho_c$, then we may similarly show that $\delta^*=-A\{\alpha_1+W\beta_1\}$.

On the other hand, N the rate of absorption of neutrons by sodium to produce sodium 24 for a fixed value of ρ_c(say ρ_0) is given by $N=(\alpha_1+\beta_1)\rho_0$. Thus

$$\frac{N}{|\delta^*|}=\frac{(\alpha_1+\beta_1)}{(\alpha_1+W\beta_1)}\frac{\rho_0}{A}.$$

Separate experiments with sodium have shown that the ratio β_1/α_1 is less than 3×10^{-2}, and since $W\simeq 0{\cdot}9$, the ratio $(\alpha_1+\beta_1)/(\alpha_1+W\beta_1)$ cannot be more than 0·3% different from unity. Therefore, in the neutron source calibration experiment, since we are only concerned with the ratio $N/|\delta^*|$, we may take $\beta_1=0$ without introducing any appreciable error.

We may thus write $\delta^*=-A\alpha_1$, $N=\alpha_1\rho_0$ and $\delta=A\{\omega S/\rho_c-B\}$ where $\omega=p(1-Y)/(1+K^2L_s^2)$.

The values of K^2 and Y have already been quoted. Measurements made on Gleep give $L_s^2=0{\cdot}0250\pm0{\cdot}0015\ \mathrm{m}^2$; $p=0{\cdot}897\pm0{\cdot}027$. Thus $\omega=0{\cdot}875$ ($\pm3\%$).

Note added in proof. Since this paper was written, it has been found that the use of a boron trifluoride chamber in the experiment caused systematic errors in the measurements of reproduction constant. These errors depended on the pile power being used, and therefore the experiment has been repeated using a different kind of chamber.

The new value obtained for the strength of the source is $9{\cdot}7\times10^6$ n/sec. The standard error remains the same as that quoted in the paper, namely $4\frac{1}{2}\%$.

Energy Levels of F-Centres

By L. PINCHERLE
Telecommunications Research Establishment, Ministry of Supply, Malvern, Worcs.

Communicated by R. A. Smith; MS. received 5th October 1950, *and in amended form 8th March* 1951

ABSTRACT. The energy levels of F- and F′-centres are calculated on a very simple model. The ground state of F-centres is found to be determined mainly by the radius of the hole that represents the missing ion in Jost's model, and to vary only slightly with the dielectric constant.

In thallium and silver halides the long-wave photoconductivity is probably due to F-centres. In PbS the energy levels of F- and F′-centres are much too deep to be responsible for photoconductivity. D-centres consisting of a pair of adjacent missing ions are also investigated : they may act as shallow traps, thereby reducing electron mobilities.

§ 1. INTRODUCTION

If a negative ion is missing in an ionic crystal, the vacant lattice site can bind one or more electrons. In univalent crystals, such as NaCl, a vacant lattice site which has trapped one electron is called an F-centre, and one which has trapped two electrons an F′-centre. In bivalent crystals we may envisage the possibility of having three electrons trapped at a lattice point, and we may call this an F″-centre. Electrons trapped at F-centres behave in a very similar way to atomic electrons, and may be represented as moving round the centre in Bohr orbits.

Many phenomena are associated with the existence of F-centres: in particular they may play a part in photoconductivity. Thus it is of interest to try and estimate their energy levels. The problem has been discussed by various authors (Tibbs 1939, Seitz 1946, Mott and Gurney 1948, Muto 1949, Simpson 1949), who have pointed out that the potential acting on even a single electron ought to be calculated by a self-consistent method. Such a method was in fact applied in a simplified form by Simpson, who carried out computations for NaCl and AgBr.*

The aim of the present paper is to point out that Simpson's results may readily be obtained by a very simple treatment. It is then reasonable to assume that the method may be applied equally well to other ionic crystals. A general formula for the energy of the ground state is obtained, which shows that this energy depends primarily on the radius R of the spherical hole in a uniform polarizable medium, which, in Jost's model, represents the missing ion. The dielectric constant of the medium has only a slight effect, and the relation $E \propto K^{-2}$ suggested by an analogy with the hydrogen atom is not even approximately verified.

The energy levels of F′-centres, and of centres consisting of a pair of adjacent missing ions (D-centres), are also discussed. Special attention is given to PbS.

---

* Since this paper was written, a different treatment of the problem has become available (Inui and Uemura 1950), in which the lattice is treated as a large symmetric molecule. It is shown that the agreement with experiment of the theoretical values for the absorption peak of the F-centres improves when one takes into consideration more and more lattice points. However, this procedure soon becomes very complicated and does not appear to converge rapidly. It is also stated, in contradiction to other investigators' conclusions, that polarization has not an appreciable effect on the energy levels.

§ 2. F-CENTRES

To obtain the potential of the field in which an electron trapped at an F-centre moves we shall use Simpson's model, that is, we shall assume that the potential is constant and equal to V_0 inside the whole sphere of radius R, where

$$V_0 = -\frac{\alpha_M Z e^2}{a} + \frac{Ze^2}{R}\left(1 - \frac{1}{K_0}\right). \qquad \ldots\ldots(1)$$

Here α_M is Madelung's constant (1·747 for cubic structures), Z is the charge of the missing ion, a is the lattice spacing (distance between nearest neighbours), and K_0 is the high-frequency dielectric constant. For the ground state, for which the probability of finding the electron outside the hole is small, we shall also assume that, for $r>R$, $V=-Ze^2/K_0r$, i.e., we shall use the 'large distance' value of the dielectric constant (Mott and Gurney 1948, p. 85) throughout. The discontinuity in the potential at $r=R$ is relatively unimportant.

The largest uncertainty is of course in the value of R. This can be calculated by a method due to Mott and Littleton (1938), and we have used only their first approximation as, for the accuracy required, it seemed unnecessary to go further: the first approximation is quite good when the polarizability of the metal ion is much smaller than that of the negative ion; it becomes worse for metals with a higher polarizability, and the results for Rb and Cs salts are therefore less reliable.

It is also debatable whether it is legitimate to take m equal to the mass of a free electron, but as the F-centres are near the bottom of the conduction band, this would appear to be a fair approximation.

The energy of the ground state of the centre can now be worked out by elementary methods. Schrödinger's equation, in atomic units, is

$$\frac{d^2\psi_1}{dr^2} + 2(E - V_0)\psi_1 = 0 \quad \text{for} \quad r<R \qquad \ldots\ldots(2)$$

and

$$\frac{d^2\psi_2}{dr^2} + 2\left(E + \frac{Z}{K_0 r}\right)\psi_2 = 0 \quad \text{for} \quad r>R$$

where ψ is normalized according to $\int_0^\infty \psi^2\,dr = 1$. Writing

$$2E = -q^2 \quad \text{and} \quad 2(E - V_0) = p^2, \qquad \ldots\ldots(3)$$

the solution for $r<R$ is $\psi_1 = A\sin pr$, $\ldots\ldots(4)$

while for $r>R$ we shall take the asymptotic solution

$$\psi_2 = Be^{-qr}r^{Z/K_0q}, \qquad \ldots\ldots(5)$$

which satisfies (2) up to terms in r^{-2}. Formulae (4) and (5) for ψ must join smoothly at $r=R$, thus:

$$p\cot pR = -q + Z/K_0qR. \qquad \ldots\ldots(6)$$

Writing $x=pR$; $y=qR$; $M_0=ZR/K_0$, equation (6) becomes

$$x\cot x = M_0/y - y, \qquad \ldots\ldots(7)$$

which must be satisfied together with the additional condition (3):

$$x^2 + y^2 = -2V_0R^2. \qquad \ldots\ldots(7')$$

The Figure shows y as function of $2V_0R^2$ for different values of M_0. The energy is then $E_{1s} = -13{\cdot}5\,y^2/R^2$ ev. As an example we consider NaCl, the substance studied also by Simpson.‡ Mott and Littleton's first approximation gives $R=0{\cdot}94a$. As $a=5{\cdot}31$ atomic units and $K_0=2{\cdot}25$, one obtains $V_0=0{\cdot}218$ A.U. Then $-2V_0R^2=10{\cdot}8$, $M_0=2{\cdot}22$. From the Figure $y=2{\cdot}45$, and hence $E=-3{\cdot}26$ ev. (Simpson finds $E=-3{\cdot}2$ ev.). Values for other substances are given in Table 1.

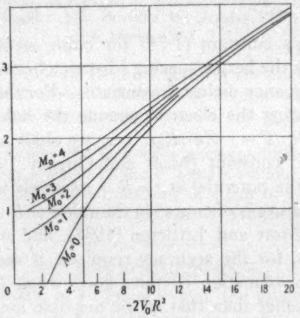


Table 1. Energy Levels of F-Centres

Crystal	$-E_{1s}$ (ev.)		$-E_{2p}$ (ev.)	$E_{2p}-E_{1s}$ (ev.)	
	Eqn. (7)	Eqn. (8)		Eqns. (7), (11)	Exptl.†
LiCl	3·21	3·30	0·91	2·30	3·1
NaCl	3·25	3·22	1·13	2·12	2·7
KCl	2·69	2·67	0·91	1·78	2·2
RbCl	2·31	2·25	0·71	1·60	2·0
CsCl	1·91	1·89	0·54	1·37	2·0
LiBr	2·90	2·73	0·80	2·10	2·7
NaBr	2·98	2·92	1·00	1·98	2·3
KBr	2·56	2·54	0·89	1·67	2·0
RbBr	2·21	2·22	0·72	1·49	1·8
CsBr	1·83	1·87	0·54	1·29	—
AgBr*	1·87	2·15	0·29	1·58	2·5

* With $R=0{\cdot}9a$. † (Ivey 1947). At 20° C.

To obtain a rough general formula we observe that, for many ionic crystals, $Z/K_0q\simeq 1$, or $M_0\simeq y$.§ Thus equation (7) may be simplified to $x\cot x=1-y$, and y is now a function of the single parameter $2V_0R^2$. We have then

$$\frac{dy}{d(-2R^2V_0)}=\frac{2R^2V_0+y}{4R^2V_0(1+y)+4y^2}\simeq\frac{1}{2(1+y)},$$

and integrating, $y=-1+(1-2R^2V_0)^{1/2}$. Hence

$$E_{1s}=-3{\cdot}78[-1+(1-R^2V_0/3{\cdot}78)^{1/2}]^2/R^2 \text{ ev.} \qquad \ldots\ldots(8)$$

where V_0 is in ev. and R in A. The energy is thus determined primarily by R. The dielectric constant enters in V_0 and also affects the ratio R/a, but its effect on E_{1s} is small, and certainly the relation $E_{1s} \propto K_0^{-2}$ suggested by an analogy with the hydrogen atom is not satisfied.

---

‡ For AgBr, also treated by this author, the value of R cannot be worked out by Mott and Littleton's method, as the polarizability of the Ag ion is not known in this substance. With the value of R used by Simpson the present method gives exactly his result.

§ This is equivalent in effect to considering the centre as a hydrogen atom, but only in the region where ψ is small.

For NaCl equation (8) gives $E_{1s} = 3{\cdot}22$ ev. In this approximation the average value of the distance of the electron from the centre is 3·6 A.U.; Simpson gives 3·0 as the mean Bohr orbit radius.

The 2p-state is the only excited state likely to be of importance for optical transitions. The peak in the absorption curve of an F-centre corresponds to the 1s–2p transition; the cut-off wavelength for photoconductivity also corresponds to this transition, the electrons being subsequently excited to the conduction band by thermal fluctuations, the rearrangement of the ions providing a large fraction of the required energy.

A 2s-state will also exist in most alkali halides. As the potential is not really spherically symmetrical, we cannot strictly say that angular momentum is quantized. In particular we cannot say that the 2s-state is metastable; but certainly the transition 1s–2s will be much less probable than the transition 1s–2p, and we shall therefore consider only the 2p-state.

For $r < R$ Schrödinger's equation is

$$\frac{d^2\psi_1}{dr^2} + \left(p^2 - \frac{2}{r^2}\right)\psi_1 = 0, \qquad \psi_1 = A\left[\frac{\sin pr}{pr} - \cos pr\right]. \qquad \ldots\ldots(9)$$

For $r > R$ we cannot now assume $V = -Ze^2/K_0 r$, as the orbit will be large compared with R; rather the dielectric constant will gradually change from the value K_0 at infinity to a value near the static one K at $r = R$. To avoid carrying out the self-consistent procedure we shall write

$$V(r) = -\frac{Ze^2}{K_0 r} + \frac{Ze^2 R}{K_0 r^2}\left(1 - \frac{K_0}{K}\right) \qquad \ldots\ldots(10)$$

and take for ψ_2 the asymptotic solution $\psi_2 = Be^{-qr}[r^{Z/K_0 q} + br^{Z/K_0 q - 1}]$, with $b = (R/2y^3)[2(M_0 - M + 1)y^2 + M_0 y - M_0^2]$, where $M = RZ/K$. The boundary conditions now lead to

$$-1 + \frac{x^2}{1 - x\cot x}$$
$$= \frac{-y^5 - (M_0 - M + 1)y^4 - (\frac{1}{2}M_0 - M + 1)y^3 + \frac{1}{2}M_0(M3_0 - 2M + 1)y^2 + M_0^2 y - \frac{1}{2}M_0^3}{y^4 + (M_0 - M + 1)y^3 + \frac{1}{2}M_0 y^2 - \frac{1}{2}M_0^2 y} \qquad \ldots\ldots(11)$$

from which the energy may be calculated without any difficulty. For NaCl the result is $E_{2p} = -1{\cdot}13$ ev., to be compared with Simpson's value $-1{\cdot}0$. Values for other substances are given in Table 1.* These results are probably high for the reasons discussed in Simpson's paper. $\overline{r_{2p}}$ for NaCl is 6·7 (Simpson's Bohr orbit = 5·6), so that also in the excited state, the orbit is not so large as it is sometimes thought to be; 92% of the charge is within two lattice distances.

If, in order to explain changes of activation energy with impurity concentration, one must assume overlapping of centres in semiconductors in which it is known that conductivity is due to F-centres, the present results indicate that the impurities must be mainly concentrated in certain regions of the crystal, or form threads through the material.

---

* The result for AgBr agrees within 10% with Simpson's, adopting his value for R.

Results may be compared with the observed peak in the absorption spectrum of the centres, which corresponds to the 1s–2p transition.† For AgBr the experimental value corresponds instead to the cut-off wavelength for photoconductivity. The calculated values for the energy difference between the 1s and 2p states are in each case about 0·7 of the experimental values; this is partly due to the fact that values of E_{2p} are almost certainly high; it may also indicate that our model treats the electron as less tightly bound that it really is. The deviation may also be due to having taken the effective mass equal to the free electron mass. In this connection it is interesting to observe that in KCl the binding energy of a positive hole at a missing positive ion site is between two and three times that of an electron of an F-centre (Dorendorf and Pick 1950). If the experimental values are extrapolated to absolute zero, by assuming $dE_{1s}/dT \simeq 6\boldsymbol{k} = 5\times10^{-4}$ ev/degree, and $dE_{2p}/dT=0$ (Mott and Gurney 1948, p. 116), they become higher by 0·1 to 0·2 ev., and the agreement becomes worse.

§ 3. F′-CENTRES

The problem of F′-centres is analogous to that of the helium atom, with a nuclear charge that may differ from 2, and is in fact equal to 1 for alkali halides.

One may take the previously determined wave functions for an F-centre and use perturbation methods. A more convenient way is to use a variational method: the model is in any case a crude one, and it will thus be sufficient to introduce the simplest possible wave function.

The Hamiltonian is

$$H = -\frac{1}{2}\frac{d^2}{dr_1^{\,2}} - \frac{1}{2}\frac{d^2}{dr_2^{\,2}} + \sum_1^2 \left\{ \begin{array}{l} V_0 \;\; (r_i<R) \\ -\dfrac{Z}{K_0 r_i} (r_i>R) \end{array} \right\} + \frac{1}{K^* r_{12}}, \qquad \ldots\ldots(12)$$

where the suffixes 1, 2 denote the two electrons and K^* indicates an average dielectric constant. We take as variational function

$$\psi = 4\gamma^3 r_1 \exp(-\gamma r_1) r_2 \exp(-\gamma r_2), \qquad \ldots\ldots(13)$$

and the energy is determined by

$$\frac{d}{d\gamma}\int \psi^* H \psi \, d\tau_1 \, d\tau_2 = 0. \qquad \ldots\ldots(14)$$

The integral involving r_{12} requires some care, as we must decide which value of the dielectric constant to use. Again a self-consistent procedure would appear necessary, but we shall adopt a simpler method by observing that the integral represents the potential energy of electron 1 in the field of electron 2. Then, when $r_2<r_1$, and the charge due to electron 2 may be considered as being at $r=0$, the high-frequency dielectric constant must be used, as electron 1 is 'at a large distance'. When $r_2>r_1$, however, and electron 1 is inside the charge considered, the static dielectric constant applies instead. In other words, at large distances from the centre the field due to any electron must be $\boldsymbol{e}/K_0 r^2$.

---

† For this comparison E_{2p} ought strictly to be the energy of that state in which the electron has a 2p wave function, but is acted upon by the field corresponding to a 1s wave function. This correction, however, is very small (Simpson 1949).

We shall thus write the last integral in (14) as

$$16\gamma^6\int_0^\infty \exp(-2\gamma r_1)r_1^2\,dr_1\left[\frac{1}{K_0 r_1}\int_0^{r_1}\exp(-2\gamma r_2)r_2^2\,dr_2 + \frac{1}{K}\int_{r_1}^\infty \exp(-2\gamma r_2)r_2\,dr_2\right] = \frac{5}{16}\gamma\left(\frac{1}{K_0}+\frac{1}{K}\right). \quad \ldots\ldots(15)$$

The variational integral is then

$$\int\psi^* H\psi\,d\tau = \gamma^2 + 2V_0 - 2\exp(-2\gamma R)\left[2R\left(RV_0+\frac{Z}{K_0}\right)\gamma^2 + \left(2RV_0+\frac{Z}{K_0}\right)\gamma + V_0\right] + \frac{5\gamma}{16}\left(\frac{1}{K_0}+\frac{1}{K}\right), \quad \ldots\ldots(16)$$

which is a minimum when γ satisfies

$$2\gamma + \frac{5}{16}\left(\frac{1}{K_0}+\frac{1}{K}\right) - 2\exp(-2\gamma R)\left[-4R^2\left(RV_0+\frac{Z}{K_0}\right)\gamma^2 + 2R\frac{Z}{K_0}\gamma + \frac{Z}{K_0}\right] = 0. \quad \ldots\ldots(17)$$

This equation is easily solved numerically.

Introducing into (16) the value of γ which satisfies (17), we find the energy E of the total system, from which we must subtract the energy E_0 of the singly ionized trap, in order to obtain the required binding energy E' of the F′-centre. The formulae of the previous sections might be applied, but for consistency it seems better to calculate also E_0 by a variational method.

Taking the same wave function as before, we have

$$E_0 = \tfrac{1}{2}\gamma_0^2 + V_0 - \exp(-2\gamma_0 R)[2R(RV_0 + Z/K_0)\gamma^2 + (2RV_0 + Z/K_0)\gamma + V_0],$$

where γ_0 is the solution of the equation:

$$\gamma_0 = \exp(-2\gamma_0 R)[-4R^2(RV_0 + Z/K_0)\gamma_0^2 + 2R(Z/K_0)\gamma_0 + Z/K_0].$$

The one-electron values given by this procedure are about 0·85 of the values given by the method of §2. Table 2 gives results for NaCl, KCl, PbS and BaO. For KCl, identifying the binding energy with the long-wave cut-off of the absorption spectrum of the centre, the theoretical value is again low. For BaO the experimental value is that of an activation energy of uncertain origin determined by thermal experiments.§ The case of PbS is discussed below. For divalent crystals, of course, the ionic picture is less applicable: moreover, in oxides an excess of the metal is known to result in interstitial positive ions and not in F-centres.

Table 2. Energy Levels of F′-Centres

Crystal	Z	γ	E (ev.)	γ_0	E_0 (ev.)	E' (ev.)	E' (exp.)
NaCl	1	0·292	−4·19	0·330	−2·91	−1·28	—
KCl	1	0·258	−3·32	0·302	−2·47	−0·85	−1·2
PbS	2	0·362	−6·04	0·375	−3·37	−2·67	(−0·28)†
BaO	2	0·364	−6·30	0·382	−3·59	−2·71	(−1·2 to −1·7)‡

† From photoconductivity cut-off (extrapolated to 0° K.).

‡ From resistance–temperature curve.

---

§ Communicated by Prof. Sproull at the Reading Conference on Semiconductors, July 1950.

§4. F-CENTRES AND PHOTOCONDUCTIVITY

It has often been suggested that the long-wave photoconductivity of many crystals is due to F-centres *. In alkali halides the phenomena connected with the absorption of light are fairly well understood; for other substances experimental data are scanty. The photoconductivity of silver and thallium compounds has been studied fairly extensively; unfortunately the present theory cannot be applied to these salts with confidence, as it was developed for strongly ionic crystals. In particular the value of R, on which the energy levels primarily depend, cannot be determined by the Mott–Littleton first approximation, as the procedure converges slowly for these substances, and in any case is not completely justifiable.

Values for AgBr were given in Table 1. Similar results are obtained for other Ag and Tl compounds. The agreement is slightly worse than for alkali halides. However, relative values are in better agreement than absolute values, so there seems to be an indication that the long-wave photoconductivity of these substances is due to F-centres. But more experimental data are necessary on materials with varying number of defects.

A substance of particular interest is PbS. The Mott–Littleton first approximation here gives $R=0{\cdot}83a=4{\cdot}65$ A.U. The static dielectric constant is 18; the high-frequency dielectric constant may be taken as 6, since this is approximately its value in the spectral region around 6μ, far from the main absorption.† The binding energy of an F′ centre, calculated by the method of §3, is then 2·67 ev. These traps are certainly too deep to account for the infra-red conductivity which cuts-off at about 0·3 ev.

There remains the possibility of a third electron being trapped at the centre (F″-centre). This electron, following the hydrogen atom analogy, would be in the 2s-state; its binding energy must then in any case be much less than that of the other electrons. Now it is easy to see that, at least for PbS, no bound 2s-state exists. In fact, if we take the ψ given by (13) for the two 1s-electrons, then, using an expression analogous to (15) to obtain the potential due to the charge distribution of these two electrons, it is immediately apparent that near the origin they reduce the absolute value of V_0 by an amount $2\gamma/K$, γ being the solution of (17). On the other hand, at large distances they completely neutralize the charge of the centre, and the potential for an additional electron is zero. We can then treat the third electron as being in a potential well of depth $V=V_0-2\gamma/K$ and radius R, the potential being zero outside. As the total quantum number is 2, the quantity $x=pR$ must be greater than $3\pi/2$. Thus $x^2>22{\cdot}2$, and, from (7′), $-2V_0R^2$ must be greater than 22·2 for a bound state to exist. Substituting numerical values, the left-hand side is approximately 11, and the formation of the F″-centre is therefore impossible. Thus in PbS and similar substances the mechanism of photoconductivity must be quite different. This is indicated also by other evidence, for instance by the fact that the cut-off shifts to longer wavelengths when the temperature is lowered.

---

* For instance by T. S. Moss (1950). Moss observes, using the K_0^{-2} law, that, if λ_c is the cut-off wavelength for photoconductivity (in μ), $\lambda_c K_0^2=$constant. The theoretical value of the constant is 10, while experimental data indicate, according to Moss, a value around 76. According to the present investigation there is no reason to expect a relation of this kind.

† This value was deduced from unpublished data of Dr. A. F. Gibson on the reflectivity of PbS in this spectral region.

§ 5. D-CENTRES

It has often been suggested that vacant sites in ionic crystals may tend to cluster (Mott and Gurney 1948, p. 41, Seitz 1946, p. 396); in particular a vacant positive-ion lattice site (bearing an effective negative charge) and a vacant negative-ion lattice site (bearing an effective positive charge) will attract each other and tend to form an electrically neutral double vacancy which we shall call a D-centre.

It is difficult to estimate how numerous these centres may be, as the activation energy for the formation of a defect is not known, except on some substances showing high ionic conductivity. Seitz estimates that in KCl at 560° c. they are as numerous as isolated defects, and that, on lowering the temperature, the D-centres may be frozen in.

These centres provide two levels equally displaced on either side of each band (if the effective masses of electrons and holes are equal). As they are neutral, they do not contribute to the conductivity; however, the level near the bottom of the conduction band may act as a shallow trap, thereby reducing the electron mobility. An estimate of the binding energy of an electron trapped at one of these centres may be made by applying the same methods (Hylleraas 1937) used in the problem of the hydrogen molecule ion, except that in our case the two 'nuclei' have opposite charge.

Let 1 be the position of the missing negative ion: we have there effectively a positive charge Z. Let 2 be the position of the missing positive ion, with effective charge $-Z$. The potential energy of an electron near the centre should be worked out by the same methods as for an F-centre. As a first approximation, however, we shall take simply a Coulomb field, modified by the polarization of the medium, i.e. $V = -Z/K_0 r_1 + Z/K_0 r_2$, where K_0 is the high-frequency dielectric constant. As the orbit of the electron is presumably rather large, this approximation seems justifiable.

In a treatment of the hydrogen molecule ion, the attraction of the two centres must also be included in the potential energy, but in a crystal we assume that the centres are held in place by the surrounding ions and not affected by the trapping of an electron. Thus there are no further terms to consider in the potential energy.

Separation of Schrödinger's equation in elliptical coordinates (prolate spheroids), $\xi = (r_1 + r_2)/a$; $\eta = (r_1 - r_2)/a$, leads, for the ground state, to the equations

$$(\xi^2 - 1)\frac{d^2X}{d\xi^2} + 2\xi\frac{dX}{d\xi} + (-\lambda^2\xi^2 + \mu)X = 0 \qquad \ldots\ldots(18)$$

and

$$(1 - \eta^2)\frac{d^2Y}{d\eta^2} - 2\eta\frac{dY}{d\eta} + (\lambda^2\eta^2 - \beta\eta - \mu)Y = 0, \qquad \ldots\ldots(19)$$

with $\lambda^2 = -\frac{1}{2}a^2E$, $\beta = 2aZ/K_0$. μ is a separation parameter. Equations (18) and (19) are to be solved under the usual conditions of continuity, etc. The η-equation provides a relation between μ and λ, and the ξ-equation determines λ, and hence the energy.

Expansion of Y in terms of the Legendre functions leads to an infinite determinant from which the relation between μ and λ may be deduced by successive approximations. For small values of β (less than 2 A.U.), we have $\mu = \frac{1}{6}\beta^2 + \lambda^2(\frac{1}{3} + \beta^2/45)$; a good approximation when β lies between 2 and 4 is

$\mu = -0{\cdot}16 + 0{\cdot}25\beta + 0{\cdot}054\beta^2 + 0{\cdot}5\lambda^2$. Only the first two terms of the development are important, and they are of the same order of magnitude. The charge density thus depends on η roughly according to a law $(1-\eta)^2$, thereby showing that the charge is mainly concentrated on the side of the positive centre, as one would expect. Writing now $X = (\xi^2 - 1)^{-1/2}\psi$; $x = \xi - 1$; $\mu = A + B\lambda^2$,

$$p(x) = -\frac{A}{x(x+2)} - \frac{1}{x^2(x+2)^2}; \quad w(x) = \frac{(x+1)^2 - B}{x(x+2)},$$

equation (18) becomes $d^2\psi/dx^2 - [p(x) + \lambda^2 w(x)]\psi = 0$. We use a variational procedure, minimizing

$$M = \int_0^\infty \left[\left(\frac{d\psi}{dx}\right)^2 + p(x)\psi^2\right] dx \text{ subject to the condition } N = \int_0^\infty w(x)\psi^2\,dx = 1,$$

the eigenvalue λ^2 playing the role of a Lagrangian multiplier. Thus we find the minimum, without conditions, of $M + \lambda^2 N$. We take as trial function $\psi = Cx\exp(-\gamma x)$. Varying then with respect to C and γ, we obtain the equations

$$M + \lambda^2 N = 0; \quad \frac{dM}{d\gamma} + \lambda^2\frac{dN}{d\gamma} = 0, \qquad \ldots\ldots(20)$$

and γ will be determined by the condition

$$M\frac{dN}{d\gamma} = N\frac{dM}{d\gamma}. \qquad \ldots\ldots(21)$$

In a Coulomb field the best approximation is then to take $\lambda = \gamma$. Carrying out the elementary integrations, equation (21) is, in full,

$$(1-B)\epsilon^2(4\gamma) + \left[\frac{3A}{2\gamma^3} + \frac{2(A+1)}{\gamma^2} + \frac{5-B}{2\gamma} + 4(1-B)\right]\epsilon(4\gamma) + \frac{2A-1}{2\gamma^3} + \frac{4A+3}{2\gamma^2} + \frac{2}{\gamma} + 3(1-B) = 0, \qquad \ldots\ldots(22)$$

where $\epsilon(4\gamma) = 4\gamma e^{4\gamma}\,\mathrm{Ei}(-4\gamma)$, the exponential integral being defined by $-\mathrm{Ei}(-x) = \int_x^\infty (e^{-t}/t)\,dt$. When γ is small, it is sufficient to keep only its lowest powers in (22); then $\epsilon(4\gamma) = -\{(3+4A)\gamma + 2A - 1\}/\{(4+4A)\gamma + 3A\}$. Over the range $0{\cdot}7 < A < 1{\cdot}3$, $\gamma \simeq 0{\cdot}314A - 0{\cdot}179$. We see that in this approximation A is the only parameter determining the eigenvalue. For $A < 0{\cdot}5$ ($b < 1{\cdot}0$ A.) there is no bound state.

Calculations were carried out for AgBr and PbS. Results are given in Table 3.

Table 3

	a (A.U.)	Z	K_0	β	μ	λ	E (ev.)
AgBr	5·44	1	4·62	2·35	$0{\cdot}73 + 0{\cdot}43\lambda^2$	0·051	−0·005
PbS	5·64	2	6	3·75	$1{\cdot}54 + 0{\cdot}50\lambda^2$	0·40	−0·27

The thermal activation energies will be smaller. These results indicate, of course, only the order of magnitude, but they show that the mobility of electrons in PbS and other semiconductors will be reduced if these centres are present. Finally we investigate whether a second electron may be trapped at a D-centre in PbS; we treat the interaction E_i between the two electrons by a perturbation method, expressing E_i by a formula similar to (15). Taking polar coordinates for simplicity, and writing (approximately) $\psi = N\cos\theta r\exp -\lambda r$, we have

$E_1=(185/1536)\lambda(1/K_0+1/K)=0{\cdot}29$ ev. for PbS. Thus the total energy of the centre with two trapped electrons is $2(-0{\cdot}27)+0{\cdot}29=-0{\cdot}25$ ev., and the binding energy of the second electron is practically zero.

ACKNOWLEDGMENTS

The author desires to thank all his colleagues at T.R.E. who helped him with many discussions. This paper is published by permission of the Chief Scientist, Ministry of Supply, and the Controller, His Majesty's Stationery Office.

REFERENCES

DORENDORF, H., and PICK, H., 1950, *Z. Phys.*, **128,** 166.

HYLLERAAS, E. A., 1937, *Z. Phys.*, **71,** 739.

INUI, T., and UEMURA, Y., 1950, *Progr. Theor. Phys.*, **5,** 252.

IVEY, H. F.,1947, *Phys. Rev.*, **72,** 341.

MOSS, T. S., 1950, *Proc. Phys. Soc.* A, **63,** 167.

MOTT, N. F., and GURNEY, R. W., 1948, *Electronic Processes in Ionic Crystals* (Oxford: University Press), chap. IV.

MOTT, N. F., and LITTLETON, M. J., 1938, *Trans. Faraday Soc.*, **34,** 485.

MUTO, T., 1949, *Progr. Theor. Phys.*, **4,** 243.

SEITZ, F., 1946, *Rev. Mod. Phys.*, **18,** 384.

SIMPSON, J. H., 1949, *Proc. Roy. Soc.* A, **197,** 269.

TIBBS, S. R., 1939, *Trans. Faraday Soc.*, **35,** 1471.

LETTERS TO THE EDITOR

On the Reciprocal Field Theory

To avoid the complications of quantum electrodynamics we explain the ideas of reciprocity on the simplest field model: a complex scalar field ψ in interaction with a real scalar field ϕ. In terms of the traditional local theory the wave functions obey the equations

$$(\Box-m^2)\psi=-g\phi\psi \quad \ldots\ldots(1') \qquad (\Box-k^2)\phi=-g\psi^*\psi. \quad \ldots\ldots(1'')$$

In conformity with Born's brilliant ideas of non-localizability and reciprocity (Born and Peng 1944, Born and Green 1949), developed further by Yukawa (1950), we assume that ψ depends upon the operators of position and displacement x_μ, d_ν: $[x_\mu, d_\nu]=\delta_{\mu\nu}$. Modifying slightly the original equations of Yukawa we postulate the following reciprocal commutation relations

$$[x_\mu,[x_\mu,\psi]]=\lambda^4[d_\mu,[d_\mu,\psi]] \qquad \ldots\ldots(2)$$

$$[d_\mu,[x_\mu,\psi]]=0, \qquad \ldots\ldots(3)$$

which are assumed to be valid even in case of an interaction present. The field ϕ is assumed to be localizable for simplicity.

In the representation where x_μ are diagonal ψ is a matrix which may be also written as a function of two variables $\psi(x, r)$ and equations (2) and (3) assume the form (Yukawa 1950)

$$r_\mu{}^2\psi(x,r)=\lambda^4\Box_x\psi(x,r) \qquad \ldots\ldots(2')$$

$$r_\mu\frac{\partial}{\partial x_\mu}\psi(x,r)=0. \qquad \ldots\ldots(3')$$

In order to establish a correspondence and an interpretation of the non-local formalism I postulated previously (Rayski 1950) that for any gauge invariant polynomial $a(x, r)$ of wave functions, formed by means of the matrix multiplication law, the observable density is

$$a(x)=\int d^4r\,a(x,r). \qquad \ldots\ldots(4)$$

For the product on the right-hand side of (1'') we get

$$v(x)=\psi^*\psi=\iint d^4r\,d^4s\,\psi^*(y,s)\psi(y,r), \qquad \ldots\ldots(5)$$

where

$$y=x-\tfrac{1}{2}(r+s). \qquad \ldots\ldots(6)$$

This enables us to write the equation corresponding to (1″) as

$$(\Box - k^2)\phi(x) = -gv(x), \qquad \ldots\ldots(7)$$

with $v(x)$ given by (5) and (6). The equation (7) may be integrated by means of retarded or advanced potentials which yields, for example,

$$\phi(x)=\phi^{\rm in}(x)+g\int dP'\Delta_k^{\rm ret}(x-x')\psi^*(y',s')\psi(y',r'), \qquad \ldots\ldots(8)$$

where $\Delta_k^{\rm ret}$ is the well-known retarded delta function for the mass constant k given by Yang and Feldman (1950) while $\int dP'$ is a short for

$$\int dP' = \int d^4x'd^4r'd^4s' = \int d^4y'd^4r'd^4s'. \qquad \ldots\ldots(9)$$

In order to write a non-local equation corresponding to (1′) we abandon, from the very outset, the infinitesimal approach through the equations (1′), (1″) and assume the following integral equation (compatible with (2) and (3))

$$\psi(y,r)=\psi^{\rm in}(y,r)+\tfrac{1}{2}g\int dP'\Delta^{\rm ret}(y-y',r,s')\{\psi(y',r'),\phi(x')\} \qquad \ldots\ldots(10)$$

constituting a straightforward generalization of the equation of Yang and Feldman (1950) adopted for the case of a scalar interaction. A similar equation holds for the outgoing waves and the advanced potentials. The ingoing (and outgoing) waves are assumed to obey the interaction-free equation

$$(\Box_x - m^2)\psi^{\rm in}(x,r)=0 \qquad \ldots\ldots(10')$$

which is reciprocal due to (2′).

The generalized $\Delta^{\rm ret}(x,r,s)$ function may be defined, together with other important Δ-like functions, in the following way: let $\hat\Delta$ denote any one of the well-known Δ, $\overline{\Delta}$, $\Delta^{(1)}$, $\Delta^{\rm ret}$ . . . functions, then its Fourier transform is $\hat\Delta(p,r,s)=\hat\Delta(p)\rho(p,r)\rho(p,s)$, where $\hat\Delta(p)$ is the Fourier transform of the usual $\hat\Delta(x)$ well known from the theory of Schwinger, while

$$\rho(p,r)=N(p)\delta(r_\mu p_\mu)\delta(r_\mu{}^2+\lambda^4p_\mu{}^2) \qquad \ldots\ldots(11)$$

where $N(p)$ is a normalizing factor such that $\int\rho(p,r)d^4r=1$. Owing to (11) we have,

$$\Box_x\hat\Delta(x,r,s)=\lambda^{-4}r_\mu{}^2\hat\Delta(x,r,s)=\lambda^{-4}s_\mu{}^2\hat\Delta(x,r,s)$$

showing that the generalized $\hat\Delta$ function is a reciprocal concept. Owing to (11), equation (10) is consistent with (2) and (3).

The equations (2), (3), (8) and (10) describe the fields ψ, ϕ including their mutual interaction in a completely reciprocal form. If the interaction vanishes then the equations (2), (3) and (10) reduce to the original equations of Yukawa with the only difference that Yukawa's λ is equal to our $m\lambda^2$.

The quantization of the reciprocal field may be achieved very simply by assuming for the ingoing and outgoing waves the following commutation relations

$$[\psi^{\rm in}(x,r),\psi^{*\,\rm in}(x',s)]=i\Delta(x-x',r,s)$$

consistent with the field equations (2′), (3′) and (10′). The commutation relations for the perturbed waves will follow automatically from the field equations (8) and (10) and may be computed by iteration.

The S-matrix of Heisenberg may be easily constructed from the integral field equations by the method described by Yang and Feldman (1950).

Thus, we possess a complete quantum formalism for the non-local scalar fields including the interaction with the full reciprocal symmetry. This formalism is free of the usual convergence difficulties.

The author is indebted to Professor M. Born for his kind interest in this work.

Nicolas Copernicus University,
Torun, Poland.
19th April 1951.

J. Rayski.

Born, M., and Green, H. S., 1949, *Proc. Roy. Soc. Edinb.* A, **92,** 470.
Born, M., and Peng, H. W., 1944, *Proc. Roy. Soc. Edinb.* A, **62,** 40.
Rayski, J., 1950, *Acta Phys. Polon.*, **10,** 103 (see also Bloch, C., 1950, *Danske Vid. Selsk.*, **26,** 1).
Yang, C. N., and Feldman, D., 1950, *Phys. Rev.*, **79,** 972.
Yukawa, H., 1950, *Phys. Rev.*, **77,** 219.

Some Remarks on the Use of β-Formalism of the Meson Field for Nuclear Interactions

Dr. A. S. Apte has pointed out that if one carries out the calculation of the decay of a vector meson into a pseudoscalar meson in the field of a nucleus employing the formalism developed in a previous paper (Vachaspati 1950, referred to below as I) one obtains a negative cross section for the process. This led to a re-examination of the formalism and is the origin of the following remarks (equation numbers refer to equations in I).

The clue to the solution of the above difficulty in Apte's results lies in the fact that Λ, defined by (3), viz. by $\beta_\mu^* = \Lambda\beta_\mu\Lambda^{-1}$, can be chosen as equal to η_0 or $-\eta_0$, depending upon whether one wants to make the Hamiltonian (14) positive definite or negative definite. Now, if the expression (14) is transformed to Proca's form in the vector meson case, one obtains a positive definite expression for H, while if a similar transformation is carried out for the scalar case, one obtains a negative definite expression, these results holding in either case independently of any special choice for Λ. Thus, in the scalar case, we get, on substituting for 1 from (S*e*)

$$\psi^\dagger\eta_0\psi = \psi^\dagger(\beta + \tfrac{1}{4}\beta_\mu\beta\beta^\mu)\eta_0\psi = \psi^\dagger(\daleth_\nu\Gamma^{\nu\dagger} + \tfrac{1}{4}\beta_\mu\daleth_\nu\Gamma^{\nu\dagger}\beta^\mu)\eta_0\psi.$$

Making use of the relation (S*e*) and employing (10) and (11), we get

$$H = \frac{\hbar c\chi}{4\pi}\psi^\dagger\eta_0\psi = -\frac{1}{4\pi}(F_0^*F_0 + (\mathbf{F}^*\mathbf{F}) + \chi^2 U^*U),$$

which is negative definite. This suggests that, in order to be consistent in our formalism, we must choose $\Lambda = \eta_0 = 1 - 2\beta_0^2$ in the vector case and $\Lambda = -\eta_0 = 2\beta_0^2 - 1$ in the scalar case. (Whenever scalar meson is referred to, pseudoscalar is also implied.) In fact, the correct procedure in the scalar case would be to start with a Lagrangian density (-1) times (12), therefrom to obtain $H = -(\hbar c\chi/4\pi)\psi^\dagger\eta_0\psi$, and then, in order to make this positive definite, to choose $\Lambda = -\eta_0$.

This difference in the choice of Λ in the vector and the scalar (or the pseudoscalar) cases gives rise to the extra minus sign necessary to make the vector → pseudoscalar decay cross section positive.

There is yet another point to be mentioned regarding the sum over polarizations carried out in (65). Writing

$$\sum_{B=1}^{10} (\beta_0)_{AB}u_{Bl} = X_{Al}, \qquad \sum_{C=1}^{10} u_{lC}^*(\beta_0)_{CB} = X_{CB}^*,$$

$$\frac{1}{2E}\left[\left\{E\beta_0 + \frac{1}{\mu}(p^m\beta_m)^2 - \mu c^2\right\}\beta_0^2\right]_{AB} = T_{AB}$$

(65) reads $\sum_{C=1}^{6} X_{Al}X_{lB}^* = T_{AB}$. Putting $A = B$, we see that the left-hand side must be positive. This means that we must have, in particular, $\sum_{A=1}^{10} T_{AA} = \mathrm{sp}\ T > 0$. However, when one actually calculates the spur of T, employing $\mathrm{sp}\ \beta_0 = 0$, $\mathrm{sp}\ (\beta_\mu\beta_\nu) = 6g_{\mu\nu}$, one finds $\mathrm{sp}\ T = -\frac{3}{E\mu}(p^2 + \mu^2c^2)$, which is negative. This shows that (65) cannot be correct.

To obtain more insight into the paradox, we remark that if we take (19) for ψ, then the statement $k_0 > 0$ is inconsistent with the facts (i) that the diagonal matrix β_0 has its first three elements equal to 1, the next three equal to -1, and the last four equal to zero, and (ii) that (22) holds. The latter discrepancy also leads to a breakdown of our interpretation that the first three solutions of ψ_A, ψ_{lA} with $l = 1, 2, 3$, correspond to positive mesons and the last three with $l = 4, 5, 6$, to negative mesons. To show this, insert (19) into the wave equation (1) and consider for simplicity the case of a meson at rest. We get

$$(k_0\beta_0 + \chi)u_l = 0, \qquad l = 1, 2, 3.$$

Employing the above representation of β_0, we get $(k_0 + \chi)u_{lA} = 0$, $A = 1, 2, 3$; $(-k_0 + \chi)u_{lB} = 0$, $B = 4, 5, 6$; $u_{lC} = 0$, $C = 7, 8, 9, 10$, $l = 1, 2, 3$.

If $k_0 > 0$, as we assume to be the case by definition, the first equation can be true only if $u_{l1} = u_{l2} = u_{l3} = 0$ $(l=1, 2, 3)$ while the last equation requires $u_{l7} = u_{l8} = u_{l9} = u_{l10} = 0$ $(l=1, 2, 3)$. Inserting these values of U_{lA} in the left-hand side of (22) we get

$$\sum_{A,B=1}^{10} u_{lA}^{*}(\beta_0)_{AB} u_{Bm} = \sum_{A,B=4}^{6} u_{lA}^{*}(\beta_0)_{AB} u_{Bm} = -\sum_{A=4}^{6} u_{lA}^{*} u_{Am} \qquad (l, m > 1, 2, 3).$$

Now, for $l=m$, this is negative. However, the right-hand side of (22) is positive since we have here restricted ourselves to values of l and m equal to 1, 2, 3 only. Thus (22) is no longer true.

The solution of the above difficulty is that we ought to have, instead of (19), for ψ the expression

$$\psi_A = \sqrt{(4\pi)}\, u_l u_{lA} \exp\,[ik_{0l}x_0 - i\mathbf{kx}]$$

with k_{0l} defined by (20) where $k_0 > 0$. When this is done (65) must be replaced by

$$\sum_{l=1}^{3} \beta_0 u_l u^{*} \beta_0 = \frac{1}{2E}\left[E\beta_0 - \frac{1}{\mu}(p^m\beta_m)^2 + \mu c^2\right]\beta_0^2.$$

If one writes $u_l^{\dagger} = u_l^{*}\Lambda = u_l^{*}\eta_0$, the above sum can be re-written as

$$\sum_{l=1}^{3} \beta_0 u_l u_l^{\dagger} \beta_0 = -\frac{1}{2E}\left[E\beta_0 - \frac{1}{\mu}(p^m\beta_m)^2 + \mu c^2\right]\beta_0^2.$$

In the scalar case on the other hand, we would get

$$\beta_0 u_1 u_1^{\dagger} \beta_0 = \frac{1}{2E}\left[E\beta_0 - \frac{1}{\mu}(t^m\beta_m)^2 + \mu c^2\right]\beta_0^2,$$

since here $\Lambda = -\eta_0$. The difference of sign in the two cases is to be carefully noted and, as remarked before, is essential to avoid the difficulty of negative probability encountered by Apte.

Institute of Theoretical Physics,
Copenhagen, Denmark.
24th March 1951.

VACHASPATI.

VACHASPATI, 1950, *Proc. Phys. Soc.* A, **63**, 1015.

The Multiple Scattering of μ-Mesons

In view of the importance of multiple scattering as a means of measuring the energy of fast particles in nuclear emulsions, an experiment has been performed to check the theoretical angular distribution using particles of known energy.

The particles chosen were μ-mesons, because it has been shown (Amaldi and Fidecaro 1950, Conversi *et al.* 1945) that their interaction with nucleons is small.

The scatterer used was a block of lead 1 inch thick, placed in the centre of a 12-inch diameter cloud chamber. Mesons were selected which traversed absorbers placed below the cloud chamber equivalent to 130 gm/cm² of lead and which came to rest in a block of carbon 3 inches thick, subsequently emitting a decay electron. Four trays of counters were used, trays 1 and 2 being above and below the cloud chamber and trays 3 and 4 above and below the carbon block. The lead absorbers which defined the energy were placed between trays 2 and 3.

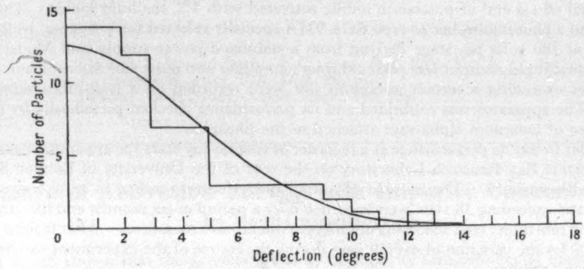
The cloud chamber was triggered by an anticoincidence (123–4) followed by a delayed pulse in tray 4 in a time interval 1·5–10 microseconds later. This method of selecting only particles which emit a decay electron identifies the particle as a μ-meson, and eliminates effects due to the inefficiency of the anticoincidence tray.

Photographs were taken with a single camera, and the projected angle of scattering was measured, the plane of projection being a plane parallel to the front glass of the chamber. The scattering angle was taken as the mean of four separate determinations. In most cases agreement between the measurements was better than 0·2 degree and in all cases it was better than 0·4 degree.

In all 72 undistorted photographs were obtained giving an average angle of deflection of 3·52 ± 0·4 degrees. The experimental histogram is shown above: the curve is a gaussian of mean value 3·52 degrees. The minimum range of mesons selected is 130 gm/cm^2 lead equivalent. This is the range of a particle which enters the apparatus vertically and comes to rest in the lower wall of the counters of tray 3. The maximum range is that of a particle which enters the apparatus at the maximum allowed angle, namely 18 degrees, and comes to rest in the upper wall of the counters of tray 4. This range is 152 gm/cm^2. From the range–energy curve (Rossi 1948) this gives the energy of the mesons selected as 184 ± 12 Mev.

According to the theory of E. J. Williams (1939) a meson of this energy would have a mean projected angle of scattering of 5·7 degrees in the 1 inch lead block.

The effect of the geometry of the apparatus on the scattering distribution has been calculated (Crewe and Litherland 1951) and shown to be very small, being only 5% at a scattering angle of 10 degrees. A detailed application of this correction shows that the mean scattering angle should be increased by 4%, giving a corrected value of 3·68 ± 0·4 degrees, that is, 65% of the theoretical value.



This result confirms the work of Kulchitsky and Latishev (1942) and also that of Sinha (1945). Using electrons of energy 2·25 Mev. and a thin lead foil Kulchitsky and Latishev found a mean angle of scattering which was 89·4% of the theoretical one, while Sinha, using mesons in the energy range 55–155 Mev., estimated that the scattering in lead was only 47% of the theoretical value.

A possible explanation of these discrepancies lies in the fact that the theory is based upon the Born approximation, and it has recently been shown (Parzen 1950) that this approximation is not valid for heavy elements. From the three experimental values quoted above, it would appear that the approximation improves as the velocity of the particles increases.

The experiment is being repeated using an iron scatterer ($Z=26$) for which the Born approximation is valid, and the theory should be correct.

George Holt Physics Laboratory,
University of Liverpool.
9th April 1951.

A. V. Crewe.

Amaldi, E., and Fidecaro, G., 1950, *Helv. phys. Acta*, **93**, 23.
Conversi, M., Pancini, E., and Piccioni, O., 1945, *Phys. Rev.*, **68**, 232.
Crewe, A. V., and Litherland, A. E., 1951, *J. Sci. Instrum.*, in the press.
Kulchitsky, L. A., and Latishev, D. G., 1942, *Phys. Rev.*, **61**, 254.
Parzen, G., 1950, *Phys. Rev.*, **80**, 261.
Rossi, B., 1948, *Rev. Mod. Phys.*, **20**, 537.
Sinha, M. S., 1945, *Phys. Rev.*, **68**, 153.
Williams, E. J., 1939, *Proc. Roy. Soc.* A, **169**, 531.

Measurements of the Star-Producing Radiation with a Scintillation Counter

A scintillation counter is being developed at Birkbeck College with the object of recording cosmic-ray stars. This type of instrument, of which numerous forms have been described in recent years, should be well suited for such an experiment since it acts as a proportional counter of small volume and high stopping power. The proportionality of the photomultiplier output to the energy lost in the phosphor, which has been established for many materials, enables us to discriminate in favour of events consisting of the simultaneous release of several heavily ionizing particles; the high stopping power and compactness of the sensitive material as compared with ionization chambers and other gas-filled devices ensures both a high rate of star production within the detector and the absence of large pulses due to showers of the soft component of the cosmic radiation.

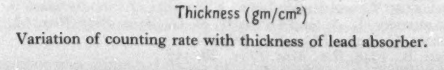
For the purpose of a preliminary investigation a counter has been constructed consisting of a crystal of 1·5 cm^3 of potassium iodide activated with 1% thallium iodide. This was attached to a photomultiplier of type RCA 931A specially selected for low noise, which was operated at 100 volts per stage derived from a stabilized power supply unit Model 1007. The photomultiplier output was passed through amplifier and pulse size discriminator units, and pulses exceeding a certain minimum size were recorded on a post-office subscriber meter. The apparatus was calibrated and its performance checked periodically by means of a source of polonium alpha-rays attached to the phosphor.

In order to test its performance as a recorder of cosmic-ray stars the apparatus was set up in the Cosmic Ray Research Laboratory on the roof of the University of London Senate House at Bloomsbury. The number of output pulses corresponding to an energy release in the crystal exceeding 38 Mev. were recorded over a period of six months and the variation of counting rate under lead absorbers of different thicknesses measured. After making small corrections for the variation of overall gain during the course of the experiment we obtained the following results:

Absorber thickness (gm/cm^2)	0	22·8	56·8	170	284
Counting rate per hour	0·250±0·017	0·232±0·022	0·198±0·022	0·164±0·014	0·118±0·009

The results are shown graphically in the Figure where the logarithm of the counting rate is plotted against the thickness in grammes per cm^2 of lead absorber.

It will be noted that the absorption is exponential. The range of the radiation recorded is 350 ± 40 gm/cm^2 of lead.



Variation of counting rate with thickness of lead absorber.

The range of the star-producing radiation in lead is known to be 310 ± 20 gm/cm^2. Comparison of the two figures strongly suggests that the events recorded by the scintillation counter are in fact cosmic-ray stars produced in the phosphor and particles from stars occurring in the surrounding material.

The rate of production of stars of three or more prongs in the unshielded phosphor at sea level is estimated as 2·5 per day. Before comparing this value with the observed rate of 6 events per day we must make allowance for the contribution of 1- and 2-prong stars and single slow protons and alpha-particles. Accurate data for such corrections are not available, but estimates based on the number of single tracks observed at sea level in Ilford C2 emulsions by Perkins (1947) and on the flux of slow protons observed at Jungfraujoch by Lattimore (1949) suggest that single tracks will account for about one event per day. An extrapolation of the formula for the integral size distribution for stars at sea level given by George and Jason (1949) suggests that a total of five counts per day may be due to stars of all sizes.

We therefore feel entitled to conclude that the observed rate agrees well with the results obtained with other methods.

The work is being continued and extended, and full details will be published in due course.

The writers' thanks are due to the London University Central Research Fund Committee with whose help the new method is being developed and to Mr. J. A. Franks of the Birkbeck College Nuffield Research Laboratory who kindly provided the crystal of activated potassium iodide.

Birkbeck College,
London, E.C.4.
12th April 1951.

E. G. Michaelis.
E. P. George.

George, E. P., and Jason, A. C., 1949, *Proc. Phys. Soc.* A, **62**, 243.
Lattimore, S., 1949, *Phil. Mag.*, **40**, 394.
Perkins, D. H., 1947, *Nature, Lond.*, **160**, 707.

Change of Activation Energy with Impurity Concentration in Semiconductors

It is well known that the activation energy of impurity semiconductors decreases with the increase of the impurity concentration. This effect was attributed by Pearson and Bardeen (1949) to a residual potential energy of attraction between the electrons (holes) and the impurity ions. According to Castellan and Seitz (1950) there is some evidence that it is the number of impurities (whether ionized or not), and not the number of free carriers, that determines the activation energy; however, it has been impossible so far to give an explanation of the phenomenon on this hypothesis, as the overlapping of the centres, assumed to be distributed uniformly, is too small to explain a relatively large effect.

It was thus thought desirable to carry out calculations of the activation energy, and of its variation with temperature, taking into account the screening of the field around a trapping centre by the free carriers, to see how far this screening might be responsible for the effect. It has been shown (Peckar 1946, Slater 1949, James 1949) that a slowly varying potential superimposed on the periodic field of a crystal may be introduced into Schrödinger's equation in order to calculate energy levels of traps and the like, provided one gives to the electron a suitable effective mass. The potential energy V of an electron in this field satisfies Poisson's equation,

$$\nabla^2 V = -(\mathbf{e}^2/\epsilon)\{n(V)-n(0)\}, \qquad \ldots\ldots(1)$$

where $n(V)$ is the free electron concentration when the potential V is present and $n(0)$ the unperturbed concentration. Equation (1) holds only at distances from the trapping centres large enough for V to vary only slightly over a lattice distance. Assuming validity of classical statistics, one may write

$$n(V) = n_0 \exp\{-(U+V)/\mathbf{k}T\}, \qquad \ldots\ldots(2)$$

where $n_0 = 2(2\pi \mathbf{m}\mathbf{k}T/\mathbf{h}^2)^{3/2}$ and $U+V$ is the depth of the Fermi level below the conduction band. If one assumes further $|V| < \mathbf{k}T$, expansion of the exponential leads to the field around a trapping centre considered as a joint charge being expressed by

$$V = (\mathbf{e}^2/\epsilon r)\exp(-r/r_0) \qquad \ldots\ldots(3)$$

with $r_0{}^2 = \epsilon \mathbf{k}Te^{U/\mathbf{k}T}/\mathbf{e}^2 n_0$. As might have been expected, r_0 is of the order of the velocity of the electrons divided by the plasma frequency.

At small distances from the centres this treatment breaks down, and it seems that statistical considerations alone are not sufficient to determine the potential. To obtain at least qualitative results one may make various hypotheses, such as: (*a*) equation (3) is valid at any distance; (*b*) write $n(V)=n(0)+(dn/dV)(0)V$ and calculate dn/dV by statistical formulae*; (*c*) fix an arbitrary upper limit to the electric density and apply (2) only so long as it does not exceed this limit. All three simplifications were tried, and they lead to similar results, at least in those cases in which the procedure is not obviously unreliable. The effective mass was taken equal to the free mass, and the uncertainty in this quantity makes a refinement of the theory rather pointless.

Calculations were carried out for n-type Si for comparison with the experimental results of Pearson and Bardeen. A self-consistent procedure must be adopted, as one must start by assuming an activation energy E in order to calculate the Fermi level; the condition is then imposed that E should coincide with the energy that one finds at the end by solving Schrödinger's equation. The dielectric constant of Si (experimentally around 13) was taken as 13·4, in order that the hydrogen atom formula should give the correct activation energy when the screening is negligible. Schrödinger's equation was solved by a one-parameter variational procedure.

The activation energies, obtained under hypothesis (*a*) above, are given in the following Table:

Activation Energy of Silicon

(1)	(2)	(3)					
		$T=50$	100	150	200	250	300
5·3	0	0·074	0·060	0·045	0·040	0·039	0·039
2·2	0·027	0·074	0·064	0·054	0·050	0·049	0·049
1·3	0·037	0·074	0·066	0·058	0·054	0·054	0·054
0·6	0·048	0·075	0·068	0·061	0·059	0·059	0·059

(1) Impurity concentration (10^{18} cm^{-3}); (2) experimental activation energy (ev.); (3) theoretical activation energy (ev.) for various values of T in °K.

The calculated values are thus higher than the experimental. As the density of free electrons is presumably above average near the positively charged centres, the simplifications adopted would tend to give values in excess. E remains fairly constant above 150° K. as found experimentally, but increases to the unscreened value at lower temperatures. No experimental evidence exists of this phenomenon, and it would be desirable to have experiments performed at temperatures lower than those investigated so far, as the quoted value of 150° K. for the beginning of the rise may well be considerably in error.

The present investigation seems then to indicate that, even if the screening of the trapping centres by the free carriers is not the only, or the main, cause of the decrease of E with impurity concentration, it does certainly reduce its value markedly.

The author desires to thank Dr. G. G. Macfarlane for helpful discussions, and Professor N. F. Mott for correspondence on the subject. This letter is published by permission of the Chief Scientist, Ministry of Supply, and of the Controller, H.M. Stationery Office.

Telecommunications Research Establishment,
Great Malvern, Worcs.
29th March 1951.

L. PINCHERLE.

CASTELLAN, G., and SEITZ, F., 1950, *Phys. Rev.*, **79**, 216.
JAMES, H. M., 1949, *Phys. Rev.*, **76**, 1602.
JAMES, H. M., and LEHMAN, G. W., 1950, *Phys. Rev.*, **79**, 216.
PEARSON, G. L., and BARDEEN, J., 1949, *Phys. Rev.*, **75**, 865.
PECKAR, J., 1946, *J. Phys. U.S.S.R.*, **10**, 431.
SLATER, N. C., 1949, *Phys. Rev.*, **76**, 1592.

---

* This approximation has been used by James and Lehman (1950) in the calculation of potential fluctuations in semiconductors.

On Mechanical Action in Dielectrics

Smith-White (1949, 1950), following Livens (1916, 1926), has recently criticized the Helmholtz theory of mechanical action in dielectrics, and has developed a theory based upon dielectric polarization. These authors appear, however, to have overlooked a subtlety which exists in the starting point of their method.

If at a point in a dielectric the electric intensity is E and the polarization P, they take the force density on the dielectric to be

$$F=(P, \text{grad})E. \qquad \ldots\ldots (1)$$

This formula does not, however, take account of the fact that a volume element at the position of E itself provides a contribution to the field of the same order as E, so that, in finding the force on the element, this contribution must be subtracted. The difficulty is of a nature similar to that experienced in determining the inducing force on a molecule in the molecular theory of dielectrics (Fowler 1936).

The matter may be investigated as follows. We consider a small element of dielectric and regard this and the rest of the dielectric as two polarized bodies. Their polarizations may then be replaced by Poisson's equivalent charge distributions. It follows that the field strength acting on the element differs from E through the removal of a volume charge of density $-\text{div } P$ belonging to the element, and the addition of a surface distribution of density P_n on the wall of the cavity occupied by the element, the suffix denoting the component of P normal to the wall and directed into the cavity. In addition to the force given by (1), one therefore has : (*a*) the force due to the charges on the wall of the cavity acting upon the charges on the surface of the element, (*b*) the forces due to the same charges acting upon the volume charge of the element, (*c*) the effect of the additional electric intensity due to the removed charge $-\text{div } P$ upon the surface charge of the element, (*d*) the effect of this electric intensity upon the volume charge of the element.

It will be easily seen that (*d*) is zero and that (*b*) and (*c*) cancel, so that all that remains is (*a*), which is, in general, not zero. The acceptance of (1) as the force density is therefore wrong.

Livens (1926) has also given an expression for the couple density in a dielectric : $G=[P, E]$. This formula is unaffected by the present criticism, for it can readily be seen that the additional field strength acting on a volume element is, with the neglect of small quantities, a multiple of P.

Smith-White is primarily concerned with the work done in a small displacement and deformation of a dielectric. In that case, since the effective extra forces are due to surface charges existing as equal and opposite adjacent layers, the total work done by them is zero. Smith-White's theory of energetics, which seems to be the most satisfactory yet evolved, is therefore justified.

It will be clear, however, that the use of (1) will not in general yield the correct result for the mechanical force on a region of dielectric, although it will in the special case where the region contains the whole of the dielectric and the dielectric is rigid, for then the additional forces cancel. In that case, however, the Livens–Smith-White theory gives the same result as the Maxwell–Helmholtz theory, as has been shown by Smith-White (1949), so that our former ideas of the mechanical action are in any case unaffected.

A quantitative study of some aspects of the general case has been made and will appear in detail elsewhere.

University College of the West Indies,
Jamaica.
20th April 1951.

R. Cade.

Fowler, R. H., 1936, *Statistical Mechanics* (Cambridge : University Press), pp. 439–444.
Livens, G. H., 1916, *Phil. Mag.*, **32,** 162; 1926, *Theory of Electricity* (Cambridge : University Press), chap. 2.
Smith-White, W. B., 1949, *Phil. Mag.*, **40,** 466; 1950, *Nature, Lond.*, **166,** 689.

Calculation of the Oscillator Strength for the $^2\Pi-^2\Delta$ Transition in the CH Molecule

In this letter the oscillator strength (f-value) for the $^2\Pi-^2\Delta$ transition in the CH molecule is calculated using the one-electron approximation (see Bates 1949, Shull 1950, Stephenson 1951). Knowledge of the f-value for this transition is of particular importance in the study of the abundance of CH molecules in interstellar space, and various estimated values have recently been used in this connection. Stromgren (1948) quotes a theoretical value of 0·002 which was obtained by Herzberg (unpublished work), whilst Dunham (1941) assumes f to be 0·06. The experimental work of Lyddane, Rogers and Roach (1941) has been corrected by Dwyer and Oldenberg (1944) and leads to an f-value of 0·0019, in good agreement with the value quoted by Stromgren.

For the transition involved, the ground state $\mathrm{X}^2\Pi$ has the configuration $\mathrm{K}(2s\sigma)^2(2\mathrm{p}\sigma)^2(2\mathrm{p}\pi)$, and the excited state $\mathrm{A}^2\Delta$ has the configuration $\mathrm{K}(2s\sigma)^2(2\mathrm{p}\sigma)(2\mathrm{p}\pi)^2$. Neglecting exchange, the transition reduces in the one-electron case to the approximate form $(2\mathrm{p}\sigma)-(2\mathrm{p}\pi)$.

The wave functions of the molecular orbitals may be approximately represented by a suitable linear combination of the atomic orbitals in the usual way.

It follows that

$$\Psi_{\mathrm{f}}(2\mathrm{p}\sigma\,|\,\mathbf{r})=\frac{\psi(2,1,0A\,|\,\mathbf{r})-\psi(2,1,0B\,|\,\mathbf{r})}{\sqrt{2}\{1-S(2,1,0)\}^{1/2}}\qquad\ldots\ldots(1)$$

$$\Psi_{\mathrm{i}}(2\mathrm{p}\pi\,|\,\mathbf{r})=\frac{\psi(2,1,\pm 1A\,|\,\mathbf{r})-\psi(2,1,\pm 1B\,|\,\mathbf{r})}{\sqrt{2}\{1-S(2,1,\pm 1)\}^{1/2}},\qquad\ldots\ldots(2)$$

where the ψ's are normalized atomic wave functions with principal, azimuthal and magnetic quantum numbers n, l, m as indicated and with the nucleus to which they apply denoted by A or B. In the present work the carbon nucleus is denoted by A.

The electronic position vector is written as $\mathbf{r}$, and the overlap integrals $S(n, l, m)$ defined by

$$S(n,l,m)=\int_\tau \psi(n,l,mA\,|\,\mathbf{r})\psi(n,l,mB\,|\,\mathbf{r})\,d\tau,\qquad\ldots\ldots(3)$$

the integration being over all space τ.

Defining the dipole moment D as

$$D=\left|\int_\tau \Psi_{\mathrm{i}}(\mathbf{r})\mathbf{r}\Psi_{\mathrm{f}}(\mathbf{r})\,d\tau\right|\qquad\ldots\ldots(4)$$

and making use of the expressions for $\Psi_{\mathrm{i}}(\mathbf{r})$ and $\Psi_{\mathrm{f}}(\mathbf{r})$ given by equations (1) and (2), it is found that

$$D=\frac{[(P+Q)-(T+R)]^2}{4\{1-S(2,1,0)\}\{1-S(2,1,\pm 1)\}},\qquad\ldots\ldots(5)$$

where

$$P=\int_\tau \psi(2,1,0A\,|\,\mathbf{r})\mathbf{r}\psi(2,1,\pm 1A\,|\,\mathbf{r})\,d\tau,\qquad Q=\int_\tau \psi(2,1,0B\,|\,\mathbf{r})\mathbf{r}\psi(2,1,\pm 1B\,|\,\mathbf{r})\,d\tau,$$

$$R=\int_\tau \psi(2,1,0A\,|\,\mathbf{r})\mathbf{r}\psi(2,1,\pm 1B\,|\,\mathbf{r})\,d\tau,\qquad T=\int_\tau \psi(2,1,0B\,|\,\mathbf{r})\mathbf{r}\psi(2,1,\pm 1A\,|\,\mathbf{r})\,d\tau.$$

The calculation of these integrals was carried out using Slater type atomic wave functions with effective nuclear charges $Z_A=3{\cdot}25$ and $Z_B=1{\cdot}00$ for the carbon and hydrogen nuclei respectively. The mean internuclear distance R was taken as $1{\cdot}118\times10^{-8}$ cm. (see Herzberg 1950). In the evaluation of the R and T integrals and of the overlap integrals it was found necessary to introduce elliptic coordinates and, in the case of the R and T integrals, to express the resulting integral in terms of Bessel functions. The values of the integrals in atomic units, the unit of length being equal to the radius of the first Bohr orbit, are : $P=0{\cdot}641$, $Q=2{\cdot}082$, $R=1{\cdot}046$, $T=1{\cdot}046$, $S(2,1,0)=0{\cdot}159$, $S(2,1,\pm 1)=0$. Using equation (5) and the expression $f=304D/\lambda$ for the f-value in terms of the dipole moment factor D (in atomic units), where λ is the wavelength in Ångström units, it is found that $D=0{\cdot}118$ and $f=0{\cdot}008$ for $\lambda=4320$ A.

Considering the approximation used, this value is in good agreement with the values given by Herzberg and by Dwyer and Oldenberg, and would seem to indicate that the value suggested by Dunham is too high.

Further calculations allowing for exchange and hybridization of the states are being considered.

Department of Physics,
Imperial College, London, S.W.7.
26th April 1951.
G. STEPHENSON.

BATES, D. R., 1949, *Proc. Roy. Soc.* A, **196**, 562.
DUNHAM, T., 1941, *Publ. Amer. Astron. Soc.*, **10**, 123.
DWYER, R. J., and OLDENBERG, O., 1944, *J. Chem. Phys.*, **12**, 351.
HERZBERG, G., 1950, *Molecular Spectra and Molecular Structure*, Vol. I: *Diatomic Molecules* (London: Macmillan).
LYDDANE, R. H., ROGERS, F. T., and ROACH, F. E., 1941, *Phys. Rev.*, **60**, 281.
SHULL, H., 1950, *Astrophys. J.*, **112**, 352.
STEPHENSON, G., 1951, *Proc. Phys. Soc.* A, **64**, 99, 425.
STROMGREN, B., 1948, *Astrophys. J.*, **108**, 242.

Superconductivity of Lead Sulphide

It has previously been reported (McLennan *et al.* 1930, Meissner *et al.* 1933, Darby *et al.* 1950) that some specimens of lead sulphide became superconducting at temperatures below 5° K. We have recently made further measurements by the induction method on two specimens of natural galena of very high purity. Both specimens were polycrystalline and p-type conductors containing respectively 7×10^{17} and 2×10^{16} charge carriers per cm^3 at room temperature*. Measurements were made at temperatures down to 1° K. and it was found that neither specimen became superconducting. In view of the negative result, it seems possible that the superconductivity observed in other specimens may be due to the presence of excess lead in the form of a thin film at the grain boundaries.

The Clarendon Laboratory,
Oxford.
15th May 1951.
J. HATTON.
B. V. ROLLIN.
E. F. W. SEYMOUR.

DARBY, J., HATTON, J., and ROLLIN, B. V., 1950, *Proc. Phys. Soc.* A, **63**, 1181.
McLENNAN, J. C., ALLEN, J. F., and WILHELM, J. O., 1930, *Trans. Roy. Soc. Can.*, **53**, 111.
MEISSNER, W., FRANZ, H., and WESTERHOFF, H., 1933, *Ann. Phys., Lpz.*, **17**, 593.

* We are greatly indebted to Dr. C. A. Hogarth of Reading University for supplying us with the specimens of galena and information about the resistivity and Hall coefficients.

On an Alternative Decay Process for the Neutral π-Meson

Observations of gamma-rays resulting from 330 MEV. X-ray bombardment of nuclei (Steinberger, Panofsky and Steller 1950) first established the existence of a neutral π-meson (π^0) and its decay into two photons. Also cosmic ray studies (Carlson, Hooper and King 1950) have shown that in large nuclear 'stars' produced in photographic plates exposed to cosmic rays, creation of neutral mesons accompanies creation of charged mesons, and a study of the energies of electron pairs found in the plates confirms the two-photon decay. Further, a study of the orientation of those pairs found related to large nuclear explosions has established an upper limit of 5×10^{-14} sec. for the decay lifetime of the neutral meson.

This two-photon decay implies spin zero for the π^0-meson (Yang 1950). The decay process may be described phenomenologically by means of the following interactions*:

$$H_{\text{int}}{}^{\text{psc}}=\eta\phi_{\text{psc}}\epsilon_{\alpha\beta\gamma\delta}\frac{\partial A_\alpha}{\partial x_\beta}\frac{\partial A_\gamma}{\partial x_\delta}=2\eta\phi_{\text{psc}}E\,.\,H, \qquad \ldots\ldots (1\,a)$$

$$H_{\text{int}}{}^{\text{sc}}=\tfrac{1}{2}\eta\phi_{\text{sc}}F_{\alpha\beta}F_{\alpha\beta}=\eta\phi_{\text{sc}}(E^2-H^2), \qquad \ldots\ldots (1\,b)$$

between the electromagnetic and neutral meson fields, according as the neutral meson is pseudoscalar or scalar. The particular mechanism of decay through the formation of a virtual proton anti-proton pair (described by Steinberger 1949) leads to these expressions (1) with particular values for η, if $(\kappa/M)^2$ is neglected in comparison with unity (κ and M are the masses of neutral meson and proton respectively). For each interaction (1) the mean life τ of the meson at rest is given by

$$\tau^{-1}=4\pi^2\eta^2\kappa^3. \qquad \ldots\ldots (2)$$

In the lifetime study of Carlson, Hooper and King (1950), the major uncertainty arose from the unpredictable deviation of the photon direction from that of the bisector of the angle between the pair particles it produces. The average pair conversion length of the photon in the photographic plate is about 5 cm. so that only a small percentage (about 3%) of the pairs resulting from the two-photon decay are available for measurement, since only pairs close (< 2 mm.) to the star are identified. Accordingly, it is of interest to note an alternative decay involving a direct pair creation, implied by the existence of the interaction (1). Its importance arises from the fact that the pair is created at the point of decay of the π^0-meson and will therefore be found close to the 'star'. In this process, the π^0-meson is transformed firstly into a real and a virtual photon, the latter then directly producing an electron pair. The ratio δ of the probability of this decay process to that of two-photon decay is, for either of the interactions (1 *a*) and (1 *b*),

$$\delta=\frac{2\boldsymbol{e}^2}{3\pi}\int_0^{\kappa^2-4\boldsymbol{m}^2}\frac{\kappa^2+2\boldsymbol{m}^2-x}{(\kappa^2-x)^2}\left(\frac{\kappa^2-4\boldsymbol{m}^2-x}{\kappa^2-x}\right)^{1/2}x^3\,dx\simeq\left(2\ln\frac{\kappa}{\boldsymbol{m}}-3\cdot5\right)2\boldsymbol{e}^2/3\pi, \qquad \ldots\ldots (3)$$

($\boldsymbol{m}$ is the electron mass), where the integrand gives essentially the energy distribution of the photons ($k=x/2\kappa$). Thus on the average 1 in 80 π^0-mesons will decay in this manner. In the meson rest system half of the pairs created directly will have angular separation θ greater than $4(\boldsymbol{m}/\kappa)^{1/2}$ i.e. greater than 15° (the distribution is approximately $d\theta/\theta$ for angles θ exceeding $8\boldsymbol{m}/\kappa$, i.e. 2°, the most frequent separation being close to 2°). Accordingly, for decay of a π^0-meson from a nuclear 'star', the angle of separation will be, on the average, somewhat larger than that between the pair electrons created by a photon from meson decay. In the published data (Carlson, Hooper and King 1950), 15 electron pairs are related to nuclear stars and of these one pair originates at the star, which is not inconsistent with this proposed interpretation for pairs observed to originate close to the star.

Observation of such direct pairs would appear to offer an independent means for the estimation of the π^0-meson lifetime, free from the particular uncertainties of the earlier method. From the (known) energy and angular distribution of neutral mesons from 'stars', and the angular distribution of the decay photons relative to the meson motion, the distribution of all pairs with distance from the star may be calculated as function of the parameter τ, the π^0-meson lifetime. For example, for π^0-mesons of momentum p and with the assumption (satisfactory for $p>\kappa$) that the photons are ejected forward from the meson, the distribution of all pairs as function of radial distance from the star is proportional to

$$\frac{e^{-r/L}-e^{-r/\lambda}}{L-\lambda}+\frac{\delta}{\lambda}e^{-r/\lambda},$$

where L is the mean conversion length for the photons ($\sim 5\times10^4$ microns) and $\lambda=p\tau/\kappa$ (with $p=2\kappa$, the most frequent case, and $\tau=T\times10^{-14}$ sec., $\lambda=6T$ microns, for example). Thus, information on the distribution of pairs close to stars could be useful for determination of τ if this exceeds 10^{-14} sec.

It may be noted here that the relation of this decay mechanism to the two-photon π^0-decay is quite analogous to the relation between 'trident' formation and bremsstrahlung along an electron path, which theory has been checked by observations in photographic plates (Hooper, King and Morrish 1951). However, a further preliminary investigation (Bristol group, private communication) of pairs within 50 microns of stars appears at present to conflict with the expectations outlined here.

---

* Natural units such that $h/2\pi=c=1$ will be used throughout.

In conclusion, I wish to thank Mr. D. T. King, who pointed out to me as a possible anomaly some pairs originating at the centre of some nuclear ' stars '.

Department of Mathematical Physics,
University of Birmingham.
10th May 1951.

R. H. Dalitz.

Carlson, A. G., Hooper, J. E., and King, D. T., 1950, *Phil. Mag.*, **41**, 701.
Hooper, J. E., King, D. T., and Morrish, A. H., 1951, *Phil. Mag.*, **42**, 304.
Steinberger, J., 1949, *Phys. Rev.*, **76**, 1180.
Steinberger, J., Panofsky, W. K. H., and Steller, J., 1950, *Phys. Rev.*, **78**, 802.
Yang, C. N., 1950, *Phys. Rev.*, **77**, 243.

REVIEWS OF BOOKS

Shorter Six-figure Mathematical Tables, by L. J. Comrie. Pp. xxvi+387. (Edinburgh and London: Chambers Ltd., 1950.) 12*s.* 6*d.*

If we divide tables of functions into two classes, the general ones and the special ones, then logarithms, trigonometrical functions, squares and square roots will certainly fall into the first class, and Hh_n functions for high n into the second. We do not want books in which a number of different special tables are collected together, but experience has shown that collections of general tables are very useful, and it is of these that we shall be speaking in the remainder of this review.

As Dr. Comrie remarks, four-figure tables are used much more widely than any others, at any rate for most scientific and engineering work, though in navigation it has been usual to use five figures and in astronomy to use seven. A very convenient set of seven-figure logarithms was issued by Messrs. Chambers in 1844 and this collection, in the form in which it was issued in 1878, became by far the most popular volume for use when four figures did not suffice. Its arrangement of trigonometrical functions was not particularly good, and it gave few of the functions, such as the hyperbolic functions, which are now in frequent use, preferring to use the space for a table of quarter-squares (which can be used for multiplication) and for traverse tables (tables of $a\cos\theta$ and $a\sin\theta$ for different values of a and of θ).

Not long ago, Dr. Comrie prepared for Messrs. Chambers a new set of six-figure tables to replace this old volume, and also a 64-page set of four-figure tables, which in a sense represented a condensation of the larger work. The volume now under review is more truly a condensation. Like the full work, it consists of six-figure tables, but with certain omissions and, in places, with increase of the tabular interval. He has now given us logarithms, natural logarithms, trigonometrical functions of angles, expressed either in radians or in degrees and minutes, the logarithms of these functions (but only for angles expressed in degrees and minutes), hyperbolic functions and powers and roots of integers. For the lower integers, these go up to the fifth power and root, and include the negative powers $-\frac{1}{2}$, -1 and -2. For the higher numbers, they stop at the fourth power and the cube root and the simple reciprocal is the only negative power. For all integers up to 1,000, the factorial and the prime factors are given, and there is a list of primes up to 12,919. There are no gamma functions.

Great care has been taken to assist the reader. The type chosen is a very legible old style, and the use of rules has been kept to a minimum, with a consequent reduction of strain on the reader's eye. First differences are given in many tables, and they are printed in italic if second differences are not negligible. Proportional parts are printed as required on the pages occupied by the logarithm table, and a complete 3×1 table of proportional parts occupies pages 358 to 377. There are also tables of interpolation coefficients in Bessel's formula.

The whole volume is very attractive, and, at its price, which corresponds to about 5*s.* before the War, should become very widely known and used.

J. H. A.

Calculating Instruments and Machines, by D. R. HARTREE. Pp. ix + 138. (Illinois: University Press, 1949; Cambridge: University Press, 1950.) 21*s*.

This book arose out of a course of lectures on computing equipment given by the author in 1948 at the University of Illinois. In an introductory chapter Professor Hartree defines his two main types of equipment. He has discarded the terms 'analogue' and 'digital' machines, which were introduced in America, in favour of the older terms, calculating instruments and machines to which they are roughly equivalent. The remainder of the book falls into two main sections which are almost independent; the first deals with computing instruments in general, and the second, with a particular section of the class of calculating machines, the automatic general purpose machines.

It is natural that the section on calculating instruments should be devoted mainly to the differential analyser since it is the only equipment in this class which has a wide range of application. In an interesting historical introduction the author shows that the fundamental ideas involved in the construction of a differential analyser were clearly appreciated by Lord Kelvin as early as 1876 though little was done towards their realization until more than fifty years later. The general structure and principles of a differential analyser are described in terms of the earlier mechanical models with particular reference to the machine installed at the University of Manchester with which the author was associated. This part of the book is particularly lucid and satisfying. An indication of the manner in which the mechanical equipment has been replaced by electrical counterparts in the newer differential analysers follows.

A second chapter on the differential analyser describes the techniques which have been developed, mainly by the author's group at Manchester University, for dealing with second order partial differential equations in two independent variables. Examples are given of the solution of parabolic partial differential equations by replacing the derivatives with respect to one of the independent variables by finite differences, and a brief comparison is made of the relative merits of making this substitution for each of the two independent variables. The use of the differential analyser for solving hyperbolic partial differential equations by the method of characteristics is described with reference to two particular problems in compressible flow, one of axially symmetric isentropic flow around a solid of revolution and the other for non-steady one dimensional flow.

The third chapter on calculating instruments gives a brief description of a number of special purpose machines including instruments for solving sets of linear equations, locating complex roots of polynomials, evaluating definite integrals and performing Fourier syntheses.

Professor Hartree introduces the section on general purpose computers with a note on terminology which should be very helpful to those to whom the subject is new. Before dealing with any specific machines he gives a fairly detailed discussion of the main features which are common to all general purpose machines. The next three chapters describe, in chronological order, the various attempts which have been made, and which are being made, to build machines to realize these objectives, from Babbage's Analytical Engine to the modern electronic high speed computers.

The chapter on Babbage's Analytical Engine should prove a useful corrective to the tendency to regard most of the basic ideas involved in the design of general purpose machines and, more particularly, in coding and programming for them, as being of recent origin. The author makes it quite clear that Babbage gave a considerable amount of thought to all these aspects of the problem, and that it is the exploitation of the recent developments in the fields of electrical and electronic engineering which chiefly accounts for the superiority of modern machines over the Analytic Engine.

The following chapter is devoted largely to a description of the Harvard Mark I machine, the first machine to realize Babbage's main objectives, and of the Eniac, the first electronic computer. The capabilities of the Eniac are illustrated by means of a problem in the theory of the laminar boundary layer in a compressible fluid which was solved by the author on this machine.

A third chapter on general purpose machines covers most of the main projects in the U.S.A. and this country at the present moment. These projects are too numerous for a detailed description to be given of each one, but a fairly clear picture of their main points of similarity and the range of variation is achieved. A section of this chapter is devoted to the use of the diagrammatic notation, introduced by von Neumann and extended by Turing, for the functional analysis of serial machines, but here the treatment suffers somewhat from overcondensation.

The book concludes with a brief review of some aspects of numerical analysis with special reference to the re-orientation which the new high speed machines will make necessary.

The author's treatment of his subject throughout is such that it will appeal to mathematical physicists rather than to those whose primary interest is in constructing equipment. The book should prove invaluable to the graduate who is interested in numerical analysis and wishes to gain a clear idea of the main trends of modern developments in this field.

J. H. W.

Textbook of General Physics, by G. R. Noakes. Pp. viii + 415. 1st Edition. (London: Macmillan and Co., 1950.) 10*s*. 6*d*.

We are entitled to expect competence in any textbook of physics at the advanced and scholarship level, but it needs something more than competence to command the wholehearted welcome which we feel this volume merits. Within the essential framework common to all who cover this ground there is ample room for teaching skill and style by which an author may succeed handsomely and deserve applause. Let us say at once that this is such a book.

The author already has volumes on light, electricity and magnetism, and heat which are widely used. He now deals with mechanics, properties of matter and wave motion. Believing that students find the mathematics difficult because they do not grasp the physical facts, he establishes his structure by appeals to experimental facts rather than by appeals to intuition—which is as it should be. This approach is reinforced by such devices as introducing the student to *Principia Mathematica* in translation and with modernized terminology and notation. The result is an excellent discussion of the laws of motion, in which, as it were, the student meets Newton in the act of discovery.

There is a welcome section on the ways in which laws may be verified (and, later perhaps, discovered) by graphical methods. There will be some readers who will think that the determination of the constants in laws of the type $P=kQ^n$ is not dealt with in the best fashion for those students who are uncomfortable with the plotting of logarithms with negative characteristics. All such difficulties vanish if the variables, P and Q, are coded by multiplication by powers of ten so as to eliminate the negative characteristics. Moreover, the procedure for fitting a straight line by least squares is arithmetically simple and might well now be taught, since simple statistics have been introduced into the Intermediate Science syllabus, presumably to give students better ability in handling experimental data. Outside the school laboratory results do not always lend themselves to fitting a line by eye.

Friction and stresses and strains are splendidly treated. It is refreshing to see a school text openly questioning whether the observed departure from Hooke's law under light loading conditions is simply due to initial kinks in the wire, and stimulating to see rubber, a very queer but essential substance, discussed on the basis of the modern work of Treloar and his colleagues. This and other quotations from current research bring home to the student that even ' elementary ' physics is still a matter for investigation.

The author's comment on the significance of terms in an equation (always a good test of an expositor's skill) can be very apt: " . . . . if a/V^2 (in the van der Waals equation) is greater still, p is zero and the container can be dispensed with, so the substance is a solid ". On the other hand, the author slips from grace, surely, when he says $p=2vmu^2/6$ is " nearer the truth " than $p=2vmu^2$ for the pressure of a gas on the wall of its container ?

There is a good section on wave motion which is rightly treated as a major topic, rather than as an incidental to sound and physical optics, and a substantial rounding off chapter on the physics of G and g.

A comprehensive range of examples from Higher Schools and Scholarship examinations is fortified by answers and, where suitable, partial solutions. Logarithmic and trigonometric tables are given. The index stood up to a stiff test, but Poisseuille's formula will be found on page 249, not 240. The production is a credit to Messrs. Macmillan as well as to the author. It is confidently recommended. M. J. M.

Physics, by S. G. STARLING and A. J. WOODALL. Pp. xvi + 1301. 1st Edition. (London: Longmans, Green, 1950.) 42*s*.

In an age of small monographs and of textbooks of composite authorship by teams of specialists this comprehensive treatise, dealing with physics in all its major branches, makes a welcome appearance. Those of us who long ago were brought up on such textbooks as Ganot, Watson, and Poynting and Thomson know full well the advantages of presentation of all parts of the subject by the same teachers whose outlook, method and style are apparent throughout: the combination we have here of an author renowned as a teacher, writer and exponent of his subject and a modern virile senior lecturer has proved particularly happy. The simple title *Physics* is the right one.

Wisely, this book is not set rigidly to any prescribed syllabus but presents basic physics, at advanced level, as seen by the authors. It will be of real value to the student reading for a general B.Sc. degree or taking a two-year ancillary course or working for special physics, particularly Part I under the new regulations of the University of London: such students will experience little trouble with the mathematics involved.

Some 250 pages are given to General Physics and Sound, while Heat and Light in 680 pages receive most generous treatment. Page 927 is reached before Electricity begins, and thus this large portion of the subject, including Atomic Physics, seems relatively cramped in some 300 pages. The early chapters on harmonic motion and gravitation are very thorough while those on elasticity, surface tension and viscosity are clear and adequate. The one conspicuous omission in this section is any treatment of moments of inertia, a subject rarely completed in an intermediate science course. The wide separation of the Sound from the General Physics is to be regretted, delaying, as it does, the chapter on wave motion (which appears long after physical optics has been treated) and necessitating consideration of forced vibration in both chapters 2 and 36.

The section on Heat is well arranged and is made unusually interesting, the chapters on kinetic theory, vapours, equations of state, solutions and the second law of thermodynamics being particularly impressive. This section closes with a chapter on statistical mechanics and quantum theory which, perhaps inevitably, seems at a level well beyond that set for the book as a whole. Jaeger and Diesselhorst's classical measurements of thermal conductivity might have had fuller treatment, showing temperature distribution expressed in terms of potential rather than distance, and one might have hoped for some detail on modern methods of measuring the mechanical equivalent of heat.

The Optics section is outstanding. The comprehensive treatment of refraction at a single surface and by a prism is good: mirrors and lenses are treated not only conventionally but by power and sagitta methods. Sign conventions are considered and frequently compared, while the useful discussions on aberrations are illustrated quantitatively by numerical data. The excellent chapters on diffraction, polarization and spectroscopy are beautifully illustrated by helpful plates and diagrams. With more care, however, given to logical arrangements space might have been saved: concepts, e.g. aplanatic surfaces, are frequently introduced when wanted and discussed much later. Angular magnification is not mentioned until the telescope is reached.

The electrical section of the book, with the exception of excellent chapters on electrolysis and magnetization of materials, seems too condensed. One might have hoped to see a short account of the Schuster magnetometer, methods of demonstrating the Peltier and Thomson effects, a vector diagram of the air transformer and an outline of the Campbell or Astbury standard of mutal inductance on which so many electrical measurements depend. The diagram (Figure 48, 8) of the N.P.L. current balance needs modification, for it is not, as stated, of the Rayleigh type but of the Ayrton–Jones pattern. Perhaps the least happy portion of the book is that dealing with alternating current bridges: from statements, repeatedly made, it is clear that the authors are under the impression that if the conditions for balance of an A.C. bridge are independent of frequency the bridge may be used with ballistic galvanometer and battery. This of course is not the true criterion of such dual use. The Owen bridge, unlike the Anderson, is completely insensitive to inductance at zero frequency and, like the Schering, when balanced ballistically requires only the fulfilment of the De Sauty relation. As the Wien bridge is given, its use for measuring power factor or frequency might have been emphasized, but a brief description of the condenser series bridge or the Schering bridge for measuring phase defect of condensers would have been more useful.

The section on Atomic Physics contains very much that is important and useful to students but appears to lack logical sequence. Under gases, electrons, etc., an article on deuterium is followed by others on the cathode-ray oscillograph, the electron microscope, time bases and radar, but electronic valves are dealt with much later under atoms and radiation in between the photo-effect and a discussion on relativity, while the magnetron appears much earlier. X-rays are treated too briefly and no modern methods of crystal analysis are given. The discovery of the neutron and its properties appear on p. 1215 but artificial radioactivity on p. 1187. Only Millikan's discarded theory of cosmic rays is given and one is led to believe that little is known about meson masses.

It is of course very easy for a specialist on any branch of the subject to suggest additions or deletions in a book of this nature or to criticize the sequence of presentation. Taken as a whole the book is really good and useful. The explanations are lucid, the type is clear and the diagrams and photographic plates, many of which have been specially prepared, are excellent. Tables of logarithms, trigonometrical functions, physical constants and principal symbols are given as well as a useful collection of examination questions which might with advantage have been dated. Reference to any part of the subject is facilitated by a well-arranged, chaptered and paged table of contents together with a good index. This portmanteau work, the best of its kind in existence, was clearly " not ordained to be gazed upon or to be carried about " but is designed for use in the home and the library. The labour entailed in its preparation has been very great and has, with a world war intervening, been spread over many years. The authors are to be congratulated on their courage in undertaking and completing this large textbook, which will be of inestimable benefit to the student and teacher.

H. R. NETTLETON.

Lehrbuch der theoretischen Physik: I—Physik der Vorgänge, by W. WEIZEL. Pp. xiv+771. (Berlin: Springer-Verlag, 1949.) In paper, 53 DM.; in cloth, 56.90 DM.

The first volume of this textbook deals with phenomenological physics, i.e. essentially with classical physics. It does not contain, however, classical statistical mechanics, which is to be treated in the second volume. It is divided into six parts. The first section contains particle mechanics and mechanics of rigid bodies, leading up to a detailed treatment of Lagrangian and Hamiltonian theories. It concludes with the transition to wave mechanics following Schrödinger's original treatment. The next section deals with hydrodynamics and elasticity. It also contains a detailed treatment of the theory of vibrations and a chapter on capillarity in addition to the fundamental principles and simple applications. The chapter on electrodynamics is subdivided in the usual way into static, stationary, quasi-stationary, etc. fields. The Maxwell equations are introduced at a fairly early stage. Optics, in addition to a fair treatment of questions of interference and diffraction, contains crystal optics at an advanced level. This is followed by a short chapter on relativity, and the book is concluded with a chapter on thermodynamics.

The book has a high standard. The presentation is clear and thorough and it follows nearly always the conventional methods of treatment. The chapter on thermodynamics should in my opinion be much longer. It also would be profitable to have a greater number of examples illustrating how to apply general principles to the solution of particular problems. The book should be very useful to the advanced student.

H. F.

Space–Time Structures, by E. SCHRÖDINGER. Pp. viii+119. (London: Cambridge University Press, 1950.) 12*s*. 6*d*.

In this little book Professor Schrödinger sets himself a limited task, namely to explain the motivation underlying recent attempts by himself and others to generalize Einstein's theory of general relativity. Demanding from the reader little previous knowledge of the subject, he develops from first principles the mathematical framework of Einstein's theory. The exposition of this theory occupies eleven out of the twelve chapters, and is arranged in such a way as to lead naturally and simply to the last chapter, which describes some of the generalizations of the theory.

The generalized field theories have been constructed as formal mathematical extensions of the Einstein theory, and their physical content (if they have any) is entirely unexplored.

Thus it is not surprising to find that the emphasis throughout this book is exclusively on pure mathematics; the physical origins and achievements of the theory are passed over with respectful silence. Like a typical modern textbook of geometry, the book starts with considerations of very wide generality and imposes successively more and more special assumptions as the development proceeds. The exposition is divided into three parts: first, the geometry of a manifold without either metric or connection; second, the geometry of a manifold without metric but with an affine connection; third, the geometry of a manifold with Riemannian metric, including the formulation of the Einstein theory. The notions of tensor, invariant differentiation, geodesic and curvature are each defined and discussed at the appropriate place, as soon as sufficiently restrictive assumptions have been made in order to give them a meaning. Building up the formalism from such an exclusively mathematical standpoint, the Einstein theory appears as a very special case at the end of the development. Just for this reason, the generalizations of the theory appear naturally as the result of dropping one or more of the special features of the Riemannian geometry, making at the same time an appropriate change in the form of the field equations.

The book is written throughout with an extraordinary lucidity, economy and liveliness of style. Within the limitations imposed by its rather narrow point of view, it is easily the clearest and most readable exposition of general relativity known to the reviewer. The author's attitude to the generalized theories, including his own, is modest and undogmatic.

F. J. DYSON.

Nuclear Physics. Notes compiled by JAY OREAR, A. H. ROSENFELD and R. A. SCHLUTER on a course given by E. FERMI. Pp. ix + 246. Revised Edition. (Chicago: University of Chicago Press, 1950.) 22*s*. 6*d*.

The compilers of this publication may be warmly congratulated for having made generally available, in the form of the reproduction of a typescript, carefully and accurately prepared lecture notes from a course on Nuclear Physics given by Professor Fermi at the University of Chicago. The scope of this course is amazing: within 240 pages it ranges from the general properties of atomic nuclei and nuclear forces to mesons and cosmic rays, and includes an account of fission and elementary pile theory. Of course, only the essentials are presented, but it is the masterly way in which this is done that confers to this course a prominent place among the numerous textbooks, some of them of great excellence, on a fashionable subject.

The course addresses itself to experimenters rather than to specialists in nuclear theory, although the latter will also greatly profit from its study on account of the sound emphasis laid everywhere on the experimental approach to the problems. Nevertheless theoretical discussions are given with all necessary details, in an admirably lucid and simple way. The book is provided with a good index, references to textbooks and papers for further study, tables of notations and constants, and a ' Segrè chart ' in which data on stable and unstable nuclei are ingeniously condensed. There is a copious supply of problems, most of which are actually suggestions for the study of interesting questions not treated in the text; the compilers have added useful references and hints to the solution. Here again the emphasis on experimental interest is apparent, sometimes in a rather overawing fashion : " Design a 10 MEV. Betatron (one week's homework)."

L. ROSENFELD.

Progress in Nuclear Physics, Vol. I, edited by O. R. FRISCH. Pp. viii + 224. (London: Butterworth–Springer Ltd., 1950.) 45*s*.

The field of nuclear physics has become so vast that nobody can possibly quarrel with the publication of surveys covering a number of disconnected areas of the field. The success of such an endeavour depends entirely on how well it is done; on the lucidity of exposition and the didactic skill with which difficult sections are presented, on the mastering by the author of the material, on the finite size, and last but not least, on the distinctness and readability of the printing of the text and formulae. It is with enthusiasm that the reviewer admits that in the present volume an excellent job has been done, and some of the chapters in fact, could hardly been have written better. The reason for this satisfactory result is obviously due to the fact that the authors are specialists in the best sense; they have apparently overcome the temptation of appearing highbrow by the simple process (loved by many theoreticians) of being unintelligible. It may, however, well be that a good deal of the result is due to Professor Frisch having taken a great deal of trouble with the editing of the text.

The first two chapters concern particle accelerators : the cyclotron by T. G. Pickavance, and the high voltage direct current generator by R. L. Fortescue. The value of the cyclotron has been enormously increased since the conception of the synchro-cyclotron and one can say that the design has, by now, probably reached a state of perfection and will not be changed very much in the future. In contrast to this, the Van de Graaff electrostatic generator, which has been developed into one of the precision tools of nuclear physics, is still a problem child when one tries to increase the voltage above 2½ million volts. The difficulties are essentially connected with the accelerating tubes, and anybody who sets out to build a machine for more than 4 million volts is well advised to look at Figure 6 on page 30 which embodies some of the disappointing experiences of accelerating tube designers. These essays are followed by a masterly survey of the photographic emulsion technique by J. Rotblat. This is a very fascinating account of how a comparatively cheap research tool can yield such an astonishing amount of new and startling information in the cosmic-ray field. All the pitfalls of the method, and the precautions to be taken when measuring tracks and the instruments required to do so, are well described.

The most readable contribution by G. G. Scarrott on electronic circuits for nuclear detectors, uses the principle of breaking down electronic paraphernalia consisting of several hundred values into simple units. This method of approach appeals to the reviewer by its novelty, and has his gratitude, as a simple-minded physicist, for its simplicity. A most lucid and complete summary of the present status of the theory of the beta-decay by T. H. R. Skyrme, and an excellent summary on the methods of beta-spectroscopy by P. E. Cavanagh, follow. Dr. Skyrme's article is particularly valuable as theoretical results are always brought into relation with experimental observations, discrepancies between the two being pointed out. Dr. Cavanagh's section deals firstly with the ' optics ' of the various beta-spectrometers, indicating (and deriving in some cases) formulae and numerical values of resolution and transmission of the various types of instruments; next, the methods of gamma-ray measurement are outlined, and finally, the arguments which permit one to determine spin changes and establish level schemes are collected.

The chapter on the Szilard–Chalmers process by H. A. C. McKay will be very welcome to physicists, as most of them will have only a vague idea about this most useful tool for preparing concentrated radioactive sources. Tables give the successful cases for neutron capture reactions and for isolating isomeric forms of active bodies. The mechanism of the process is discussed and some details for applications are indicated. The volume concludes with a review of the scattering of neutrons by J. M. Cassels. This large field is covered in 30 pages; the relevant theory is derived *ab initio* in so far as it is possible in a review and is carried through to include, finally, a solid consisting of different nuclei gifted with spin. The account of the experimental results follows the exposition of the theory; this section seems a bit short, though the author did not consider scattering by ferromagnetics and the resulting polarization effects to belong to the theme of his review. An author and subject index concludes the volume.

In view of the success of this first volume, our expectations for the second are very high indeed.

E. BRETSCHER.

The Acceleration of Particles to High Energies. Pp. x + 58. Physics in Industry Series. (London: Institute of Physics, 1950.) 10*s*. 6*d*.

An unprecedented increase in the numbers of men and in the amounts of money devoted to physical research has occurred in the last few years. This leads to a great acceleration of progress in research but has a serious consequence in that textbooks become more and more out of date. As this process continues, and the new data become more voluminous, the work and time required for a new textbook increase, and its useful life decreases. In several fields of physics the incentive to write new books has therefore fallen below the ' critical size '. Frequent surveys of rapidly advancing fields are badly needed and the volume under review is an excellent example of what can be done in a limited field. It consists of a set of papers on the main types of accelerator now of interest to physicists. The papers are essentially those given at a Conference held by the Institute of Physics in June 1949, and a report of the original discussion is included.

There are four main articles : on the cyclotron by Professor Rotblat; on betatrons and synchrotrons by Mr. F. K. Goward; on electrostatic generators and on linear accelerators by Mr. E. S. Shire. The list is thus not quite complete, no mention being made of the familiar H.T. rectifier set or of the microtron, but this is of little disadvantage; the attempt to obtain completeness is one of the factors which keep textbooks out-of-date.

Each article is extremely readable and Professor Rotblat especially has been very successful in explaining the physics of the motions of particles with only the simplest of mathematics. The value of each article is increased by the presence of the others; the best type of accelerator for any particular purpose is much easier to decide from a set of simultaneous surveys such as this than from miscellaneous original papers written at different times.

It might be noted that Veksler's first paper (in Russian) on the principle of the synchrocyclotron and the synchrotron was published in 1944, and not in 1945.

The reviewer has no hesitation in recommending the book to any physicist interested in this rapidly advancing field. J. H. FREMLIN.

Electronics, by P. PARKER. Pp. viii + 1050. 1st Edition. (London: Edward Arnold, 1950.) 50*s*.

" This book is based on lectures given . . . to undergraduate engineers, and to graduates in engineering and pure science."

This lengthy work essays to cover a rather large field in considerable detail and on the whole must be credited as a very worthy attempt, particularly useful for the engineer who will be less inclined than the physicist to cavil at any fundamental inaccuracies. The difficulty for an author in trying to deal adequately with the whole range of electronic physics and engineering—valve fundamentals, amplifiers, oscillators, circuitry, cathode-ray tubes, gas discharges, photo-cells and noise—is that, of course, of being really expert in every branch of the subject-technology as well as fundamental principles. Mr. Parker has clearly read very widely including much of the original literature, but there are some notable omissions in certain fields.

On the purely credit side I would compliment the author on his system of notes and references at the end of each chapter where less obvious points in the text are discussed in more detail, additional mathematical analyses are effected, and references to the literature are collected. Some exercises, generally of a fairly elementary character, are also provided, with answers. The presentation, general lay-out of the text and printing are very pleasing and the provision of a set of data tables, including general physical constants, comparative periodic table data, etc., is most valuable and rather unusual in such a textbook.

However, the serious student of natural science must be careful and critical when fundamental physics is discussed. A rather glaring example calling for criticism occurs in the Appendix dealing with kinetic theory and quantum statistics; the author discusses the application of Fermi–Dirac statistics to electrons in a metal but continues to apply these to *gas molecules*, such as hydrogen and helium, and ascribes the " fall in the specific heat of a gas at low temperatures " to the degeneracy of F. D. S. statistics.

In a survey today of atomic structure it seems rather a pity to present diagrams (particularly Figure 13) based on the 'old' quantum theory, the more so when it leads one to depict the inner ' S ' electrons as going round the nucleus in circles.

It must also be admitted that considerable bewilderment is felt when, in a text of this kind, one meets the statement (p. 569): " The theory (of gas discharge) speaks of ions, atoms and electrons, and of collisions between them; *but these are figments of the mind, props for its understanding* " (my italics). Furthermore such a stand-point seems to contrast rather oddly with the following extract (p. 325) . . . " We speak for convenience of the input resistance of the valve, but that is a mathematical figment, and cannot dissipate heat." One is rather left wondering to what kind of analysis the author is prepared to ascribe reality.

One cannot as a physicist dismiss inaccuracies and inconsistencies of such a nature as trivial but nevertheless the book will be of considerable value to many teaching and working in this field if it is read with reasonable discretion. D. K. C. MACDONALD.

The Identification of Molecular Spectra, by R. W. B. Pearse and A. G. Gaydon. 2nd Edition. Pp. xi + 276. (London: Chapman and Hall, 1950.) 50*s*.

The aim of this book is to guide the practical spectroscopist in correctly and completely assigning the bands in a composite spectrum to the molecules or radicles producing them. Users of the first edition, published in 1941, are unlikely to have made wrong assignments of the kind often found in earlier papers; for instance, an investigator would erroneously ascribe to a new emitter bands which he did not recognize as part of a known band-system produced by some impurity in the source.

Although several of the books on band spectra contain tables of observational data and reproductions of spectrograms which afford considerable help in this direction they are not designed for this purpose, their main theme being the structure of bands and band systems in terms of quantum theory. This book, on the other hand, is admirably planned for the purpose and is the only one of its kind. It consists mainly of two sections: (i) a table of data for about 1,800 persistent band-heads in order of wavelength, and (ii) a set of lists of data for band-systems in alphabetical order of the molecules, neutral or ionized, to which they belong. Comparison of the data for an unrecognized band-system with the data in (i) at once yields a clue to the identity of the system and the molecule; then, reference to the data for that system in (ii) indicates the presence or absence of individual bands of the system. This process of double reference is now repeated for other systems of the same molecule and of any other molecule having an atom in common with it, e.g. having identified a system of C_2 and one of N_2 we must seek not only other systems of C_2 and N_2 but also systems of CN, CH, NH, CO and NO since a trace of hydrogen or air may be present in the source. Confirmation may then be sought in an appended table of about 400 persistent atomic lines of elements arranged in alphabetical order and in the plates of excellently reproduced spectrograms.

The second edition includes new material from papers published up to 1947 and some additional references and data which were inserted in the course of proof-reading as late as November 1949. The growth of this material in recent years (and hence the need for a second edition) is evident from the fact that new band-systems or other new particulars are now given for about 90 of the 254 diatomic molecules covered by the first edition as well as for 30 others for which no published data were available ten years ago. The number of triatomic and polyatomic molecules included has risen from 34 to 47, although some for which the earlier results have now been disproved are not now included. The expansion of the tables and accompanying notes forming the text amounts to 55 pages, and four new plates (26 spectrograms) have been added to the eight of the first edition.

The new edition is a valuable complement to other books on molecular spectra and should quickly become an essential item in the observational equipment of all laboratories where spectroscopy is advanced or applied. W. J.

Diffusion Moléculaire de la Lumière et Effet Raman. (Colloques Internationaux du C.N.R.S., VII.) Pp. 145. (Paris: Centre National de la Recherche Scientifique, 1950). 27*s*.

In April 1948, a colloquium with the above title was organized at Bordeaux by Professors Kastler and Rousset, partly to mark the twentieth anniversary of the discovery of the Raman effect. As a result, many workers in this field attended. Sir C. V. Raman and Professor Max Born were present, as well as workers from Britain and the U.S.A., Austria, Belgium, Holland, and many in France.

This report of the proceedings takes the form of a series of papers, and constitutes two succeeding numbers of the *Journal de Chimie Physique* (1949, **46,** 1–2 and 3–4). Vigorous expressions of contrasting points of view were made by the two principal speakers, concerning the dynamics of crystal lattices. Other papers concerned more particularly experimental results in studies of the Raman effect and X-rays applied to crystals, especially diamond, alkali halides and similar materials and certain organic crystals. Several papers were concerned with Rayleigh and Brillouin scattering and in all there were 28 papers read. Their appearance in this form has been delayed by printing difficulties, but they are not yet 'dated', and workers in this field are recommended to obtain them, either in this form or as copies of the *Journal*, since many of the articles are important. A. C. M.

Semiconductors, by D. A. Wright. (Methuen's Monographs on Physical Subjects.) Pp. viii + 130. (London: Methuen, 1950.) 7*s*. 6*d*.

After two introductory chapters on electrons in metals and in crystals (50 pages) the author deals in the six following chapters with applications. Two of them (20 pages) are connected with problems of conductivity, the rest (78 pages) are devoted to emission of electrons from free surfaces. These latter chapters are well written and lead the reader up to recent developments; they contain a fair number of references to the original literature. Other aspects of semiconductors are, in comparison, treated in very little detail. This holds in particular of the basic questions such as structure, mean free path and others. No derivation is given for the formula describing the temperature dependence of the number of conduction electrons, whereas the question of the penetration of electrons through potential barriers is treated in some detail. It appears that the author intended the book to be mainly an introduction to the emission of electrons from surfaces. He has well succeeded in this respect. It would have been of benefit to the prospective reader if this restriction in the scope of the book had been indicated by an appropriate subtitle. H. FRÖHLICH.

Advances in Radiochemistry, by E. Broda. (Cambridge Monographs on Physics.) Pp. x + 152. (Cambridge: University Press, 1950.) 15*s*.

This admirable book by Dr. Broda is a worthy addition to the series of the Cambridge Monographs on Physics; it gives an excellent review of recent advances in selected topics of radiochemistry, as well as an account of the preparation of radio-elements by neutron irradiation.

Radiochemistry is defined by the author as the chemistry of bodies which are detected through their nuclear radiations. In a monograph of this size it is not possible to consider all the advances which have been made in this broad field. However, Dr. Broda has succeeded very well in reviewing the progress made in the last fifteen or twenty years in many of the topics of interest to the radiochemist.

The author first considers the special behaviour of systems in which the radioactive material is present in the small amount usually described as a tracer quantity. Chapters on the distribution of tracer quantities between solids and liquids, tracer quantities in gases, and the electrochemistry of radio-elements are presented. Then follows a chapter on the production of radio-elements by nuclear synthesis. This is devoted almost exclusively to neutron-induced reactions. The author considers in some detail the methods for the production of neutrons, the slowing down of neutrons and the use of slow neutrons in nuclear reactions. The last four chapters are devoted to considerations of the chemical aspects of nuclear fission, new radio-elements of special interest, specific radiochemical effects due to chemical excitation following nuclear reactions, and new developments in the technique of radiochemical measurement.

This book is not intended as a manual on radiochemistry or on its techniques. The author has assumed a knowledge of the basic facts of radioactivity and has omitted a detailed description of the standard laboratory techniques of radiochemistry. He does not consider either applied radiochemistry, i.e. the use of radioactive tracers as a tool in research investigations, or the field now generally termed radiation chemistry. These topics are considered to be beyond the scope of this monograph.

Dr. Broda has produced a well-written, concise account of recent research in the field to which he himself has made many contributions. The specialist and non-specialist alike will find in this book an admirable summary of many of the important advances which have been made in this subject. K. J. McCALLUM.

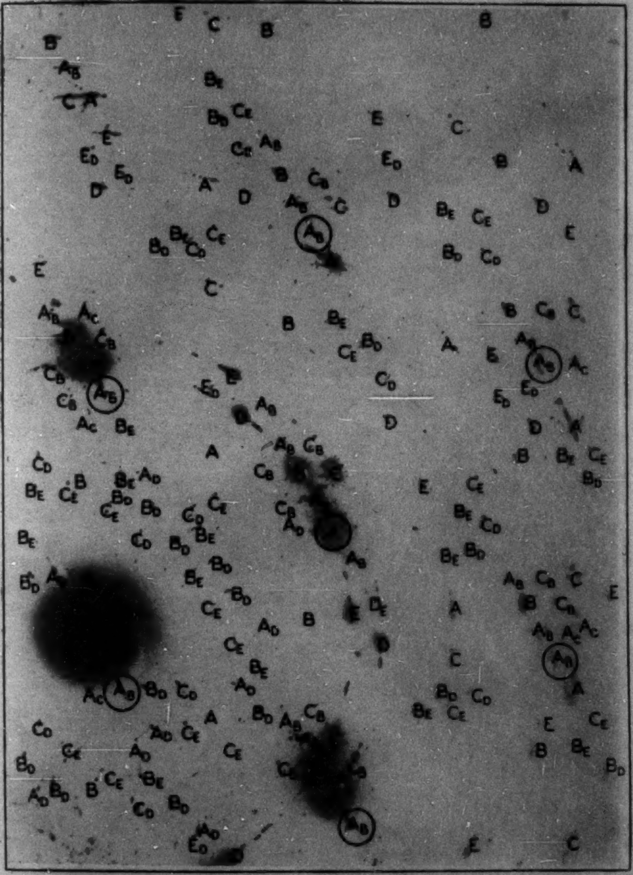

E
C
B
B
B
A_B
C A
B_E
B_D C_E
E
C
E
C_E A_B
E_D
E_D
B
A
E_D
B
C_B
D
A
D
A_B
C
D
B_E C_E
D
A_B
C_E
E
B_E
B_D C_D
B_D C_D
E
C
A_B A_C
B C_B C
B
B_E
C_B
C_E B_D
A
A_B
E
A_B A_C
C_B
C_D
E
E_D
E_D
E_D
A_B
C_B
A_B
A_C B_E
D
D
A
A_B C_B
A
B B_E C_E
C_D
C_B
E
B_E B B_E A_D
B_D
E
C_E
C_E B_D
C_E B_D
C_D C_E
B_E
C_B
A_D
B_E C_D
B_E C_D B_D B_E
B_E B_D
A_B
B_D
B_D A
B_E
A_B C_D C
E
B_D
C_B
D_E
A
C_E
A_D
B
A_B A_C A_C
C_E
D
C
A_B
B_E
A_D
A
A_C A_B B_D C_D
B_D C_D
B_E C_E
C_B
A B_D A_B
C_E
C_D A_D C_E
E
C_E
A_D
C_E
B B_E
B_D
C_E
B_D
C_E B_E
A_D B_D B
C_D B_D
A_B
A_D
E_D
E
C

Figure 1. Pattern from paraffin wax. In the transparent overlay, the five primary cross-gratings are indicated by the letters A, B, C, D, E, and pairs of letters show the origin of the secondary reflections.

Semiconductors, by D. A. WRIGHT. (Methuen's Monographs on Physical Subjects.) Pp. viii + 130. (London: Methuen, 1950.) 7*s.* 6*d.*

After two introductory chapters on electrons in metals and in crystals (50 pages) the author deals in the six following chapters with applications. Two of them (20 pages) are connected with problems of conductivity, the rest (78 pages) are devoted to emission of electrons from free surfaces. These latter chapters are well written and lead the reader up to recent developments; they contain a fair number of references to the original literature. Other aspects of semiconductors are, in comparison, treated in very little detail. This holds in particular of the basic questions such as structure, mean free path and others. No derivation is given for the formula describing the temperature dependence of the number of conduction electrons, whereas the question of the penetration of electrons through potential barriers is treated in some detail. It appears that the author intended the book to be mainly an introduction to the emission of electrons from surfaces. He has well succeeded in this respect. It would have been of benefit to the prospective reader if this restriction in the scope of the book had been indicated by an appropriate subtitle.

H. FRÖHLICH.

Advances in Radiochemistry, by E. BRODA. (Cambridge Monographs on Physics.) Pp. x + 152. (Cambridge: University Press, 1950.) 15*s.*

This admirable book by Dr. Broda is a worthy addition to the series of the Cambridge Monographs on Physics; it gives an excellent review of recent advances in selected topics of radiochemistry, as well as an account of the preparation of radio-elements by neutron irradiation.

Radiochemistry is defined by the author as the chemistry of bodies which are detected through their nuclear radiations. In a monograph of this size it is not possible to consider all the advances which have been made in this broad field. However, Dr. Broda has succeeded very well in reviewing the progress made in the last fifteen to twenty years in many of the topics of interest to the radiochemist.

The author first considers the special behaviour of systems in which the radioactive material is present in the small amount usually described as a tracer quantity. Chapters on the distribution of tracer quantities between solids and liquids, tracer quantities in gases, and the electrochemistry of radio-elements are presented. Then follows a chapter on the production of radio-elements by nuclear synthesis. This is devoted almost exclusively to neutron-induced reactions. The author considers in some detail the methods for the production of neutrons, the slowing down of neutrons and the use of slow neutrons in nuclear reactions. The last four chapters are devoted to considerations of the chemical aspects of nuclear fission, new radio-elements of special interest, specific radiochemical effects due to chemical excitation following nuclear reactions, and new developments in the technique of radiochemical measurement.

This book is not intended as a manual on radiochemistry or its techniques. The author has assumed a knowledge of the basic facts of radioactivity and has omitted a detailed description of the standard laboratory techniques of radiochemistry. He does not consider either applied radiochemistry, i.e. the use of radioactive tracers as a tool in research investigations, or the field now generally termed radiation chemistry. These topics are considered to be beyond the scope of this monograph.

Dr. Broda has produced a well-written, concise account of recent research in the field to which he himself has made many contributions. The specialist and non-specialist alike will find in this book an admirable summary of many of the important advances which have been made in this subject.

K. J. McCALLUM.

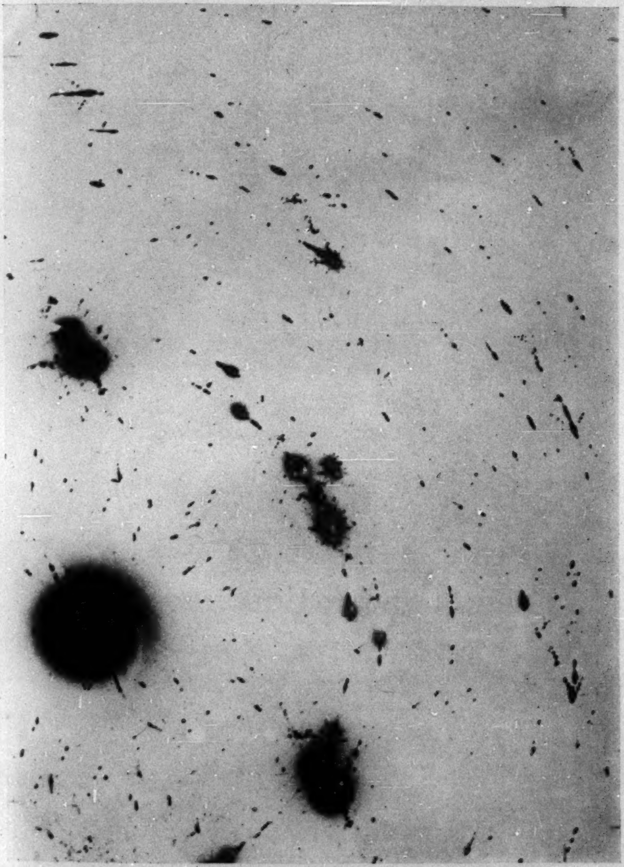


Figure 1. Pattern from paraffin wax. In the transparent overlay, the five primary cross-gratings are indicated by the letters A, B, C, D, E, and pairs of letters show the origin of the secondary reflections.

Figure 2 (*a*). Enlargement of a spot group from Figure 1.

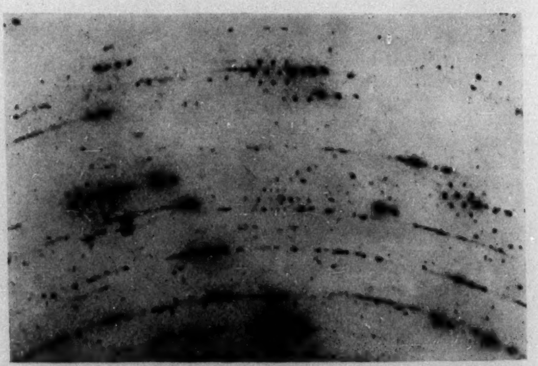


Figure 3. Regular groups of spots in a pattern from dicetyl.

PLATE II.

PROC. PHYS. SOC. VOL. 64, PT. 7—A (J. M. COWLEY, A. L. G. REES AND J. A. SPINK)

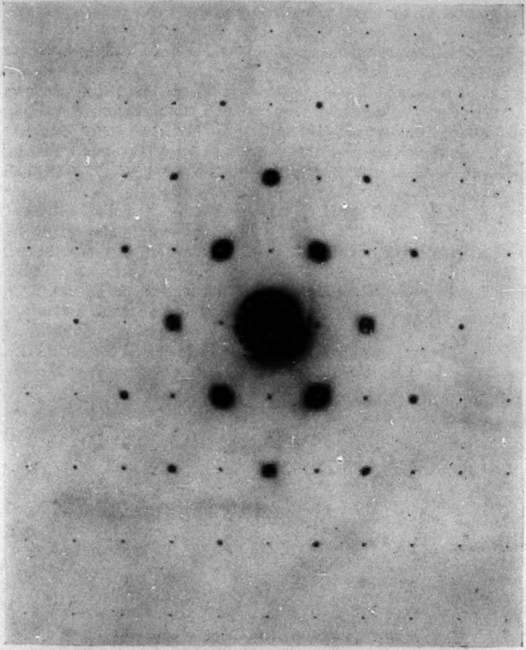


Figure 4. Single-crystal cross-grating pattern from dicetyl.

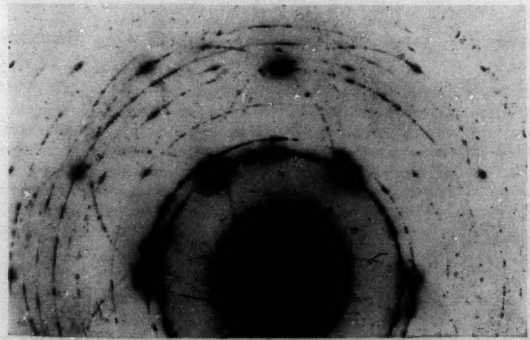


Figure 5. Pattern from paraffin wax showing complete secondary rings centred on strong primary spots.

PLATE III.

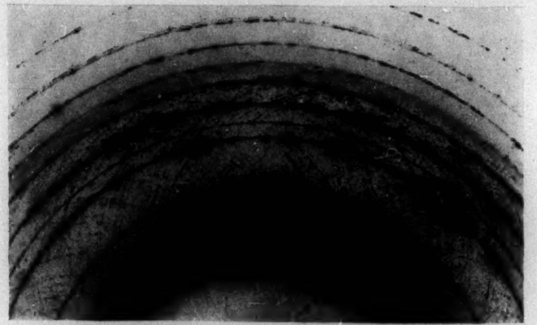


Figure 6. Portion of a paraffin wax pattern showing many secondary rings and illustrating the formation of ‘extra-ring’ envelopes.

CONTENTS FOR SECTION B

PAGE
Dr. W. D. Wright. Modern Problems of Colorimetry. (17th Thomas Young Oration.) 537
Dr. K. Hoselitz and Dr. M. McCaig. Torque Curves and other Magnetic Properties of Alcomax 549
Prof. F. Llewellyn Jones and Dr. G. D. Morgan. High-Frequency Discharges : I—Breakdown Mechanism and Similarity Relationship 560
Prof. F. Llewellyn Jones and Dr. G. D. Morgan. High-Frequency Discharges : II—Similarity Relationship for Minimum Maintenance Potentials . . . 574
Mr. S. E. Barden. On Resonance Damping at Injection in Betatrons and Synchrotrons 579
Dr. J. M. Cassels, Mr. J. M. Dickson and Dr. J. Howlett. Multiple Traversal of Cyclotron Targets 590
Mr. R. P. Chasmar and Dr. A. F. Gibson. The Characteristics of Long Period Photo-Effects in Lead Sulphide 595
Dr. A. F. Gibson. The Sensitivity and Response Time of Lead Sulphide Photoconductive Cells 603

Letters to the Editor :

Mr. J. M. Dickson. Multiple Traversal of Cyclotron Targets 615
Dr. E. H. Putley and Mr. J. B. Arthur. Lead Sulphide—An Intrinsic Semiconductor 616
Dr. P. C. Thonemann and Mr. W. T. Cowhig. The Role of the Self Magnetic Field in High Current Gas Discharges 618
Mr. P. Ravenhill and Dr. J. D. Craggs. Afterglows in Hydrogen Discharges 618
Mr. I. G. Ross and Dr. R. A. Sack. Solvent Effects in Dipole Moment Measurements 619
Corrigenda 620
Reviews of Books 620
Contents for Section A 623
Abstracts for Section A 623

ABSTRACTS FOR SECTION B

Modern Problems of Colorimetry, by W. D. Wright. (*17th Thomas Young Oration.*)

Torque Curves and other Magnetic Properties of Alcomax, by K. Hoselitz and M. McCaig.

ABSTRACT. Using a torque magnetometer, the crystal anisotropy constants of the permanent magnet alloy Alcomax III have been determined. The type of magnetocrystalline energy after rapid cooling is cubic with a positive constant (iron-like); after slow cooling cubic with a negative constant (nickel-like) and after cooling in a magnetic field uniaxial with a positive constant (cobalt-like). The changes in crystal anisotropy are compared with parallel measurements on the hysteresis curve. A direct experiment shows that when the field applied during cooling makes an angle with the crystal axes, the preferred direction is much nearer to a [100] axis than the field direction.

High-Frequency Discharges: I—Breakdown Mechanism and Similarity Relationship, by F. Llewellyn Jones and G. D. Morgan.

ABSTRACT. High-frequency breakdown between wires and coaxial cylinders in air and in hydrogen was investigated over the range of frequencies f from 3·5 Mc/s. to 70 Mc/s. at pressures below 20 mm. Hg. It was found that geometrically similar systems broke down at the same potential provided that both parameters ap and f/p were invariant, a being a

linear dimension. Under conditions such that the total amplitude of oscillation of the electron cloud was smaller than the electrode interspace, the electric field at the wire was the controlling factor, and breakdown was found to be independent of the nature of the surface of the cylinder. Consequently, secondary ionization processes did not appear to play an important part as in the case of static discharges. The experiments were in agreement with the view that the high-frequency discharge in air and in hydrogen is determined by primary ionization in the gas and loss of electrons to the walls by diffusion only, provided the electron mean free path is small compared with the linear dimension of the discharge tube, and the collision frequency is very much greater than the oscillation frequency. The significance of the similarity relation is briefly discussed.

High-Frequency Discharges: II—Similarity Relationship for Minimum Maintenance Potentials, by F. LLEWELLYN JONES and G. D. MORGAN.

ABSTRACT. The same similarity relationship which applies to breakdown applies also to the maintenance of H.F. currents, but the dependence of the minimum maintenance potential upon the parameters ap and f/p (linear dimension a, pressure p, frequency f) is not so well defined, an effect due to discharge instability.

On Resonance Damping at Injection in Betatrons and Synchrotrons, by S. E. BARDEN.

ABSTRACT. It is shown that under special conditions of injection, damping of electron oscillations can occur in 'imperfect' cyclic accelerators, i.e. those with an azimuthally inhomogeneous electromagnetic field, in the presence of resonance between their different periodic motions. This mechanism is invoked to explain the successful injection of electrons in most betatrons and synchrotrons. The necessary coupling inhomogeneity to induce resonance damping at injection can be created by the large currents circulating during this period in the acceleration cycle.

Multiple Traversal of Cyclotron Targets, by J. M. CASSELS, J. M. DICKSON AND J. HOWLETT.

ABSTRACT. In high energy cyclotrons the accelerated particles may pass through the target more than once. The average number of traversals is calculated theoretically and compared with experimental results. Good agreement is found.

The Characteristics of Long Period Photo-Effects in Lead Sulphide, by R. P. CHASMAR and A. F. GIBSON.

ABSTRACT. A photoconductive effect having a decay time of several hours has been observed in specially prepared layers of lead sulphide when maintained at low temperatures. These effects are characteristic of layers that have been treated in a certain manner with sulphur or with oxygen.

The general properties of the layers showing this photo-effect have been investigated. It is found that there are no first order differences between the properties of the layers containing oxygen and those containing sulphur. It is suggested that the phenomena are associated with the excitation of electrons from the upper filled band of the lead sulphide and the trapping of these electrons probably on the surface of crystallites. The depth of the traps is estimated to be of the order of 0·3 ev. below the bottom of the conduction band.

The Sensitivity and Response Time of Lead Sulphide Photoconductive Cells, by A. F. GIBSON.

ABSTRACT. Measurements of the response time and photosensitivity of lead sulphide photoconductive cells have been made with a view to establishing a theoretical working model for photoconductors of this type. The measurements include the variation in response time and photosensitivity with temperature, background illumination, applied electric field and other parameters. The results support, at least qualitatively, a theory of photoconductivity which postulates the existence of space-charge barriers at intercrystalline contacts. The height of such barriers is reduced on illumination and conduction facilitated.